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# SO2 Abatement for Coal-Fired Boilers in Japan

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# $SO_2$ Abatement for Coal-fired Boilers in Japan

by

Jumpei Ando

Faculty of Science and Engineering
Chuo University
Tokyo, Japan

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Project Officer

J. David Mobley
Emissions Effluent Technology Branch
Industrial Environmental Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

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The report is a compilation of information on the current status of SO2 abatement technologies for coal-fired boilers in Japan, where strict ambient air quality standards for SO2 and NOx mandate the use of various air pollution control technologies. It focuses on flue gas desulfurization (FGD) and is based on information gathered from utility company representatives and FGD process developers and the author's research in the field. Various technologies including wet lime/limestone and indirect lime/limestone FGD processes and combined FGD/Selective Catalytic Reduction (SCR) systems are described in terms of process descriptions, development and/or commercial application status, and economics. Detailed operating data are given for many of the processes.

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# ABSTRACT

Strict ambient air quality standards for sulfur dioxide ($SO_2$) and nitrogen oxides ($NO_x$) in Japan mandate the use of various air pollution control technologies. This report is a compilation of information on the current status of $SO_2$ abatement technologies for coal-fired boilers in Japan. It focuses on flue gas desulfurization (FGD) and is based on information gathered from utility company representatives and FGD process developers as well as the author's research in the field. Various technologies including wet lime/limestone and indirect lime/limestone FGD processes and combined FGD/Selective Catalytic Reduction (SCR) systems are described in terms of process descriptions, development and/or commercial application status and economics. Detailed operation data are given for many of the processes.

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# SUMMARY

The total capacity of coal-fired utility boilers in Japan is now only 4,300 MW (3.7% of total utility power) and is expected to reach 10,000 MW (5.6%) in 1985 and 22,000 MW (10%) in 1990. Several coal-fired industrial boilers may be also constructed. All of these boilers will apply flue gas desulfurization (FGD).

FGD has been widely used in Japan and is considered an accomplished technology. Over 1,200 commercial FGD units for various gas sources with various processes have been operated. FGD processes treat a total capacity of over 120,000,000 $Nm^3$/hr (40,000 MW equivalent) to remove 85-95% of $SO_2$. The principal by-products are gypsum, sodium sulfite, and sulfuric acid.

For coal-fired boilers, the wet limestone process with salable by-product gypsum has been used almost exclusively. One unit uses carbide lime and produces throw-away sludge. All of the FGD units have been operated with over 99% reliability. Since their operation is reliable, these FGD units do not have standby scrubbers.

Almost all of the new FGD units for coal-fired boilers constructed since 1978 or currently under construction have a prescrubber with a separate liquor loop to remove impurities in flue gas such as fly ash, chloride, and fluoride. They also employ low pressure drop scrubbers and a rotating gas-gas heater (heat exchanger) for gas reheating and for water conservation. In 1981 the investment cost of a new FGD limestone-gypsum process unit for a new 300-700 MW coal-fired boiler was approximately 30,000 - 35,000 yen/kW including the gas-gas heater. The annualized FGD cost is a little less than 2 yen/kW hr including 7 years depreciation.

Several modified lime/limestone processes have been developed with the goals of improving ease of operation and lowering costs. Some of the processes will be applied commercially for coal-fired boilers. Dry processes using activated carbon have also been tested. A demonstration unit of a carbon process producing elemental sulfur will be constructed in 1984.

Selective catalytic reduction (SCR) of $NO_x$ for coal-fired boilers is now being applied commercially. Most new coal-fired boilers will be equipped with an SCR reactor for about 80% $NO_x$ removal. The reactor is located between the boiler and the air preheater and operates at 350-400°C. The flue gas leaving the SCR reactor contains 1-5 ppm ammonia ($NH_3$), which is removed by the wet FGD system. The ammonia causes no adverse effects on lime/ limestone FGD processes, except that the wastewater from the FGD system contains a small amount of $NH_3$. At some of the plants, $NH_3$ in wastewater is removed by conventional activated sludge or ammonia stripping processes. Where SCR is applied upstream, the concentration of nitrate in wastewater derived from $NO_x$ in flue gas may be reduced.

Gasification, liquefaction, and fluidized-bed combustion (FBC) of coal have been tested but may not be advantageous in Japan for several reasons. Liquefaction and gasification are energy consuming and therefore too costly because Japan has to import coal. FBC processes with limestone to remove $SO_2$ create ash disposal problems. Moreover, $SO_x$ and $NO_x$ reduction by FBC may not be sufficient to meet the stringent Japanese regulations.

# SECTION 1

# AIR POLLUTION CONTROL AND COAL USAGE IN JAPAN

## 1.1 AIR POLLUTION CONTROL IN JAPAN

### 1.1.1 $SO_x$ and $NO_x$ Regulations

The ambient air quality standards for $SO_x$ and $NO_x$ shown in Table 1-1 are more stringent than those in the United States and West Germany. In order to meet the standards, $SO_x$ and $NO_x$ emissions have been regulated in the following 4 ways.

(1) Emission standards (by Central Government or Environment Agency)

(2) Total emission regulations (by Prefectural Government guided by Environment Agency)

(3) Ordinance (by Prefectures and major cities)

(4) Local agreement (between Prefectural, or City Government, or citizens group and industry)

The $SO_x$ emission standard is based on the k-value control. The specific allowable quantity of $SO_x$ emissions for each stack is calculated using the following equation:

$Q = k \times 10^{-3} He^2$

Q: Allowable $SO_x$ ($Nm^3/hr$)

He: Effective height of stack (m)

TABLE 1-1. AMBIENT AIR QUALITY STANDARDS

| Country | $SO_2$, ppm |  | $NO_2$, ppm |  |
|---|---|---|---|---|
|  | Daily Average | Yearly Average | Daily Average | Yearly Average |
| Japan | 0.04 | (0.02) | 0.04-0.06 | (0.02-0.03) |
| U. S. A. |  | 0.03 |  | 0.05 |
| West Germany |  | 0.05 |  | 0.05 |

The k-value depends on the number of $SO_x$ sources in a region and is smaller in regions with larger numbers of $SO_x$ sources. The $NO_x$ emission standards for stationary sources are based on the advanced technology of combustion modification.

The total emission regulation controls the total emissions from all factories in a city or region. It has been applied in 24 cities and industrial regions for $SO_x$ and will be applied in 3 cities for $NO_x$.

In addition, local governments and citizens groups make agreements with owners of major emission sources to restrict emissions. Usually the most stringent controls are made by such agreements. Examples of actual $SO_x$ concentrations in flue gases from coal-fired utility boilers which satisfy such agreements are shown in Table 1-2. Table 1-3 shows examples of $NO_x$ concentrations for coal-fired utility boilers. Boilers in large cities (Isogo, EPDC) and new boilers (Tomato-Atsuma, Hokkaido Electric) have the most stringent emission controls.

In order to ensure the abatement of pollutants, local governments have conducted extensive monitoring of both emissions and ambient air quality using automatic analyzer - telemeter - computer systems.

### 1.1.2 Sulfur Abatement History and Problems

Flue gas desulfurization as well as desulfurization of heavy oil, the major fuel used in Japan, started in the late 1960s. Operation problems

TABLE 1-2. EXAMPLES OF CONTROLLED $SO_2$ EMISSIONS FROM COAL-FIRED UTILITY BOILERS

| Company | Station | New or Retrofit | Capacity (MW) | $SO_2$, ppm FGD in. | $SO_2$, ppm FGD out. | Removal (%) |
|---|---|---|---|---|---|---|
| EPDC[a] | Isogo[b] | R | 265 | 450 | 20 | 95.5 |
| EPDC | Takasago | R | 250 | 1500 | 130 | 91.3 |
| EPDC | Takehara | R | 250 | 1700 | 100 | 94.1 |
| EPDC | Matsushima[c] | N | 500 | 1000 | 100 | 90.1 |
| Chugoku Electric | Shimonoseki | R | 175 | 1000 | 100 | 90.0 |
| Hokkaido Electric | Tomato-Atsuma[d] | N | 350 | 230 | 20 | 91.3 |

[a] Electric Power Development Co.
[b] In a big city
[c] Far from cities. Three-fourths of the gas is subjected to FGD.
[d] Half of the gas is subjected to FGD.

TABLE 1-3. EXAMPLES OF CONTROLLED $NO_x$ EMISSIONS FROM UTILITY COAL-FIRED BOILERS

| Power Company | Station | New or Retrofit | Capacity (MW) | $NO_x$, ppm | | |
|---|---|---|---|---|---|---|
| | | | | Emission Standard | Local Agreement | Actual |
| EPDC[a] | Isogo[b] | R | 265 | 400 | 159[c] | 180[d] |
| EPDC | Matsushima[e] | N | 500 | 400 | 300 | 250[d] |
| Chugoku Electric | Shimoniseki | R | 175 | 400 | 200 | 190[f] |
| Hokkaido Electric | Tomato-Atsuma | N | 350 | 400 | 170 | 160[g] |

[a] Electric Power Development Co.
[b] Located in urban area
[c] Requested by local government: the figure is under negotiation
[d] Attained by combustion modification
[e] Located in rural area
[f] By selective catalytic reduction
[g] By combustion modification and selective catalytic reduction

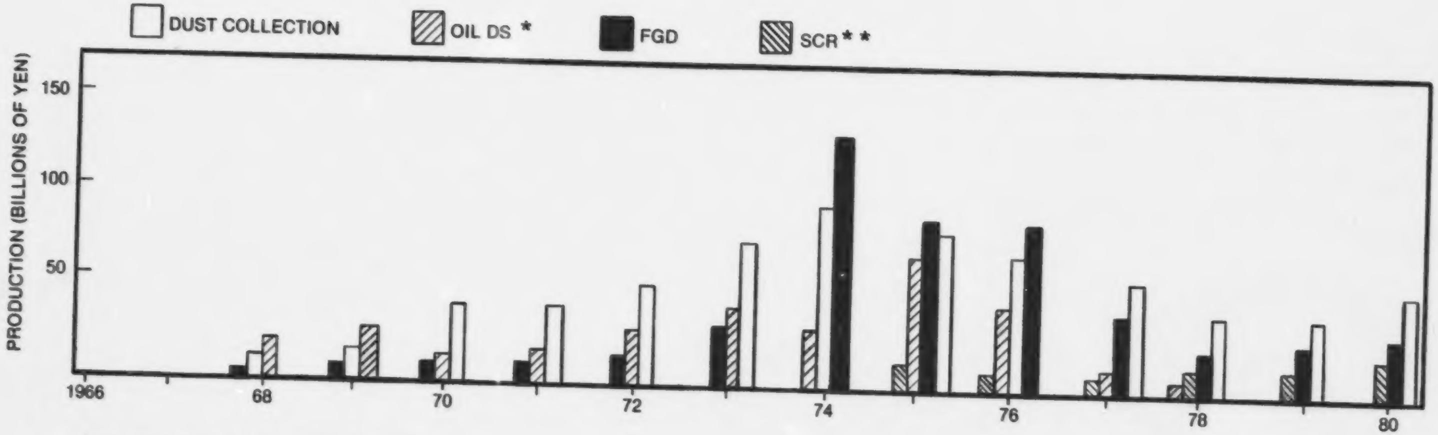
in early FGD systems forced shutdown of the gas sources because the regulations, enforced by extensive monitoring, did not allow violations. Given this situation, both process developers and users worked to improve the FGD processes, and they have succeeded in establishing reliable technologies.

As shown in Figure 1-1, the number of FGD plants started to increase rapidly in 1972, and the yearly production reached a maximum value of 150 billion yen in 1974. The total number of FGD plants has exceeded 1,200 with a total capacity of nearly 50,000 MW equivalent. Many plants for hydrodesulfurization of fuel oil were also constructed. The yearly production of the hydrodesulfurization plants reached a maximum of 75 billion yen in 1975.

There were five factors that contributed to the construction of many desulfurization plants: (1) stringent regulations and extensive monitoring, (2) establishment of technologies, (3) government assistance through the provision of low-interest loans for the construction of pollution control facilities, (4) rapid industrial growth between 1965 and 1975 which enabled industry to invest extensively for pollution control, and (5) shortage of sulfur and its compounds which has made FGD by-products valuable.

Due to desulfurization efforts and the importation of low-sulfur fuels, ambient $SO_2$ concentrations have decreased remarkably since 1967 to meet the stringent ambient air quality standard (Figure 1-2). Yearly production of desulfurization facilities decreased after 1976 because of lower ambient $SO_2$ concentrations, economic depression, and an oversupply of FGD by-products. The yearly production of FGD plants dropped to 20 billion yen in 1978 and then began to increase gradually because of recent increases in the use of high-sulfur oil and coal due to the shortage of low-sulfur oil.

Another driving force for $SO_2$ abatement has been the $SO_2$ levy applied in designated regions. Since 1974 levies have been applied to $SO_x$ sources with exhaust gas rates greater than 5,000 $Nm^3/hr$ (1.7 MW equivalent) in some regions and 10,000 $Nm^3/hr$ (3.4 MW equivalent) in others (Table 1-4). The owners of the $SO_2$ source must pay the levy according to the amount of $SO_2$



*OIL DS: HEAVY OIL DESULFURIZATION

**SCR: SELECTIVE CATALYTIC REDUCTION OF $NO_x$

Figure 1-1 Annual Production of Air Pollution Equipment.

70A2470

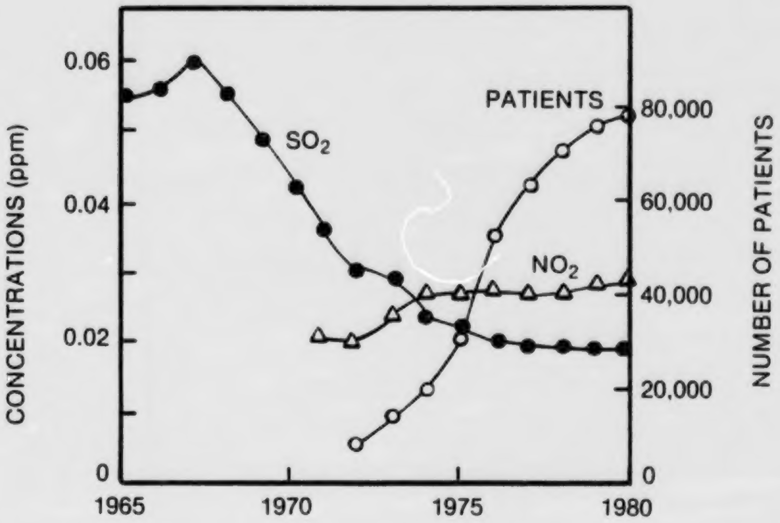


**Figure 1-2** Average Annual $SO_2$ Concentrations in 15 Major Cities and Industrial Regions and Number of Designated Air Pollution Patients.

emitted, even if the emissions are low enough to meet the regulations. The levy has been used to aid certain air pollution patients in designated regions. The patients must be diagnosed by an assigned medical doctor as air pollution victims. The designated regions initially included only heavily industrialized regions, but they have been expanded to include large parts of major cities such as Tokyo and Osaka. This expansion produced a significant increase in the number of designated patients, even though $SO_2$ concentrations decreased (Figure 1-2). As a result, the levy rate (yen/$Nm^3$ of $SO_2$) has increased tremendously as shown in Table 1-4. Some of the industries have installed FGD systems to reduce the levy, although their $SO_2$ emissions are low enough to meet the regulations. Such efforts, however, only increase the levy rate, as long as the number of the patients does not decrease. The total $SO_2$ levies reached 50 billion yen in 1980.

TABLE 1-4. $SO_2$ LEVIES (YEN/$Nm^3$ of $SO_2$)

| Regions | 1974 | 1975 | 1976 | 1977 | 1978 | 1979 | 1980 | 1981 |
|---|---|---|---|---|---|---|---|---|
| Osaka | 16 | 37 | 210 | 536 | 774 | 1239 | 1515 | 1564 |
| Tokyo | 16 | 37 | 210 | 381 | 430 | 646 | 871 | 923 |
| Chiba | 16 | 37 | 210 | 345 | 344 | 517 | 644 | 722 |
| Omuta | 16 | 37 | 210 | 307 | 301 | 452 | 492 | 522 |
| Other | 2 | 9 | 23 | 34 | 48 | 72 | 84 | 89 |

This situation has caused political problems. Industry claims that because the ambient $SO_2$ concentration has decreased, the designated regions should be reduced so that the number of designated patients is reduced. Conversely, others claim that the increase in the number of patients has been caused by $NO_x$ and want more stringent regulations on $NO_x$, including $NO_x$ levies. They also want to designate $NO_x$-rich regions as polluted regions so that the number of the patients will further increase. The number of patients leveled off in 1980-1981.

### 1.1.3 Sulfate Concentrations

Figure 1-3 shows the change in annual average concentrations of air pollutants in the five most polluted cities--Tokyo, Osaka, Kawasaki, Amagasaki, and Kitakyushu. Sulfate concentrations (sulfuric acid and its compounds) increased between 1968 and 1970, decreased substantially between 1970 and 1973, and have since increased slightly, while $SO_2$ has continued to decrease since 1968.

There are two major origins of sulfuric acid mists or sulfates. One is $SO_2$, which is oxidized in the atmosphere in the presence of radiation, moisture etc., to form fine particulates of dilute sulfuric acid. This causes a visible haze in the air as well as acid rain. The pH of the acid rain may be as low as 3 but the concentration of the acid is very low--0.005% $H_2SO_4$. On the other hand, there is a large amount of fine mist present. In Japan, there have been substantial increases in visibility and decreases in the amount of acid rain with the decrease in ambient $SO_2$ concentrations.

The other major origin of sulfuric acid mist is $SO_3$ which is contained in small amounts in combustion gases as well as in the waste gases of chemical plants, including those which produce sulfuric acid. The $SO_3/SO_2$ ratio of combustion gases normally ranges from 0.01 to 0.05. $SO_3$ combines with moisture to form mists of concentrated sulfuric acid (70-100% $H_2SO_4$). The amount of sulfuric acid derived from $SO_3$ may be less than that derived from $SO_2$ but mists of concentrated sulfuric acid can be more hazardous to human health, unless the $SO_3$ is emitted from a high stack. When it is released from the stack, the mist's acid is diluted by moisture absorption and air before humans are exposed to it.

The increase in ambient sulfate concentrations between 1968 and 1970 may be due to a rapid increase in the consumption of heavy oil, which often has replaced coal as a fuel (Figure 1-4). Although flue gases from heavy oil and coal both contain $SO_2$ and $SO_3$, the $SO_3/SO_2$ ratio is about 3/100 for the oil and 1/100 for coal. Heavy oil also contains an appreciable amount of vanadium which is emitted as fine particulates in flue gas. As shown in Figure 1-3, the vanadium concentration in ambient air also increased between 1968 and 1970.

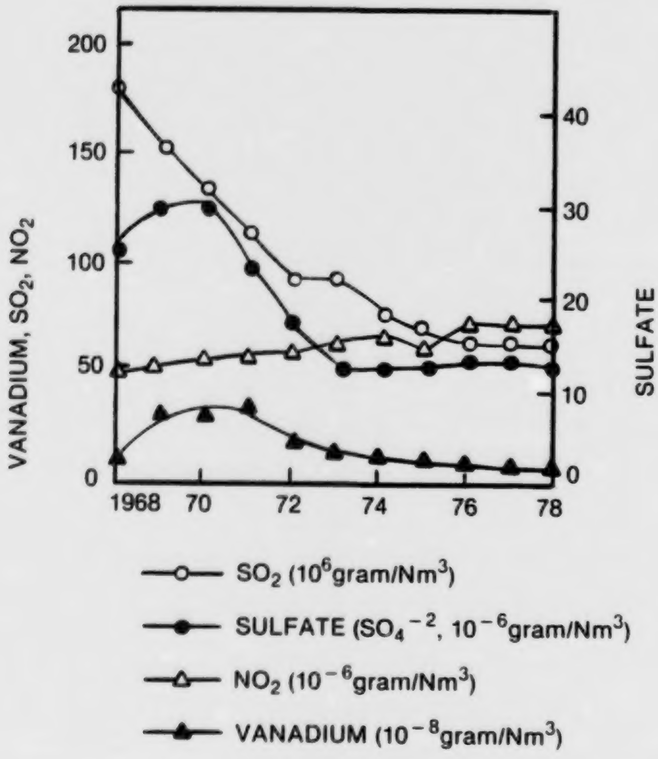


**Figure 1-3** Annual Average Concentrations of Pollutants in 5 Major Cities.

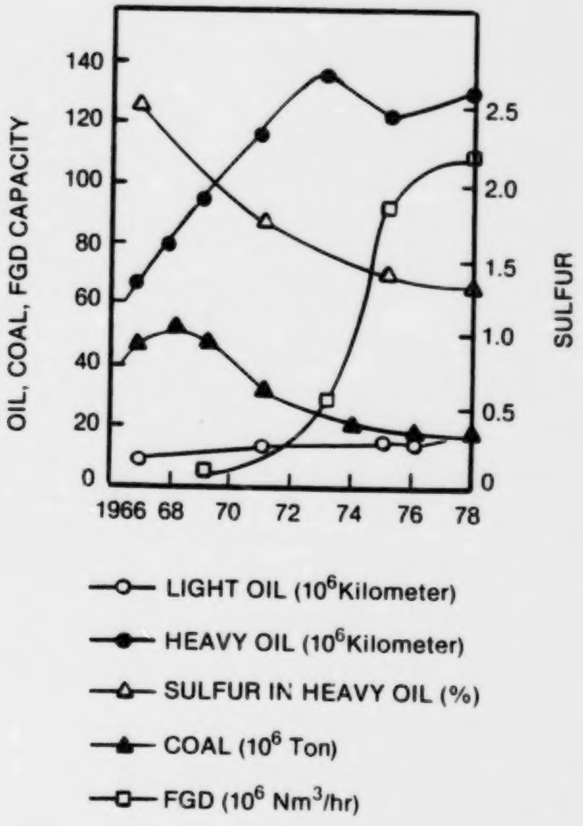


**Figure 1-4** Fuel Consumption, Sulfur in Heavy Oil and FGD Capacity.

Similar phenomena occurred in the U.S. between 1963 and 1969. Ambient $SO_2$ levels decreased in many regions, but sulfate did not decrease significantly and even increased in some regions (1). An increase in vanadium was also observed (Figure 1-5). These changes also may have been caused, at least in part, by the increased use of low-sulfur oil replacing coal during this period. Vanadium in the atmosphere may also serve as a catalyst to promote the oxidation of $SO_2$.

In Japan, both sulfate and vanadium concentrations in ambient air decreased between 1970 and 1973. This decrease may have been be due to the following reasons: 1) development of FGD which removes about 90% of $SO_2$ together with about 70% of particulates including vanadium and also about 50% of sulfate, 2) an increase in the use of electrostatic precipitators (ESP) to remove particulates including vanadium from oil-fired boilers, and 3) ammonia injection in oil-fired boiler flue gas to remove sulfuric acid mist which is converted to ammonium sulfate and caught by ESP.

Since 1973, ambient $SO_2$ and vanadium concentrations have decreased with the increasing development of FGD while sulfate concentrations have increased slightly. The increase in sulfate might be due to the increase in diesel engine vehicles, mainly buses and trucks, which use light oil containing about 0.4% sulfur (Figure 1-4).

In 1977 the average ambient sulfate concentration in the major cities in Japan was about 15 μg/Nm³ and did not exceed the California air quality standard of 25 μg/Nm³. Presumably a considerable portion of the 15 μg was due to sulfuric acid mists in diesel engine car exhausts. These mists are hazardous because of the danger of human exposure to the high concentration of sulfuric acid. Analysis of the air along roads and highways showed that concentrated sulfuric acid mists with particle sizes ranging from 0.1 to 100 μm existed along with carbonacious particles which also contain concentrated sulfuric acid (2). In addition, carbon works as a catalyst to convert $SO_2$ to sulfuric acid. The amount of concentrated sulfuric acid mist in cities may be much higher in Japan than in the U.S. because of the higher sulfur content of light oil as well as the larger number of buses and trucks in

Japanese cities. It may be necessary to reduce the sulfur content of the light oil and to improve diesel engines in order to reduce particulates, including acid mists.

The formation of $SO_3$ in combustion gases can be reduced by combustion with low excess air, which also reduces $NO_x$ formation. Wet FGD processes usually remove up to about 50% of $SO_3$ together with about 90% of $SO_2$. Some dry FGD processes may remove less $SO_2$ but more $SO_3$. In Japan, a wet electrostatic precipitator is now being used downstream of wet FGD. This system removes virtually all of the particulates including $SO_3$ or sulfuric acid mists. Baghouses (fabric filters) downstream of FGD also are useful for reducing these mists.

## 1.2 COAL USAGE AND POLLUTION CONTROL

### 1.2.1 Coal Consumption and Associated Problems

Most utility companies in Japan switched fuel from coal to oil between 1960 and 1974. One exception was the Electric Power Development Co. (EPDC) which was established by the Japanese government jointly with major power companies to use domestic coal. Due to the recent rise in oil and gas prices, however, power companies have begun to construct new coal-fired boilers again. Most of these boilers will use imported coal since the supply of domestic coal is limited to 20 million tons a year. All of the 60 million tons of coal that Japan has been importing annually have been used for coke production by the steel industry. Now Japan also imports fuel coal and is expected to import as much as 54 million tons of it in 1990 (Table 1-5). The total capacity of coal-fired boilers was 4300 MW (3.7% of total utility power) in 1979 and is expected to increase to 22,000 MW (10%) in 1990 (Figure 1-6).

The major problems associated with coal usage are: 1) emissions of $SO_2$, $NO_x$ and particulates during combustion, 2) handling and storage problems, and 3) ash disposal. Those problems are serious in Japan where a

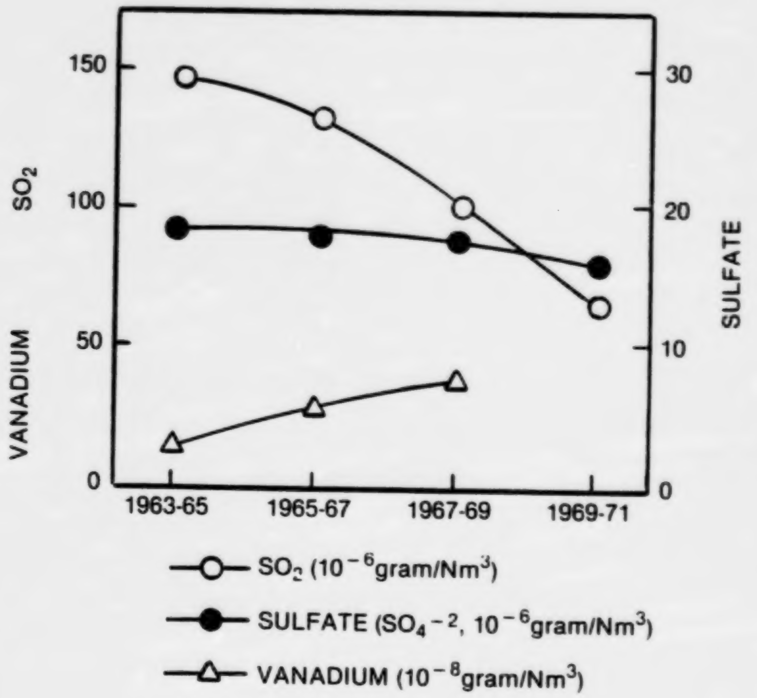


**Figure 1-5** Ambient Pollutant Concentrations on the East Coast of the U.S.: 1963-1971.

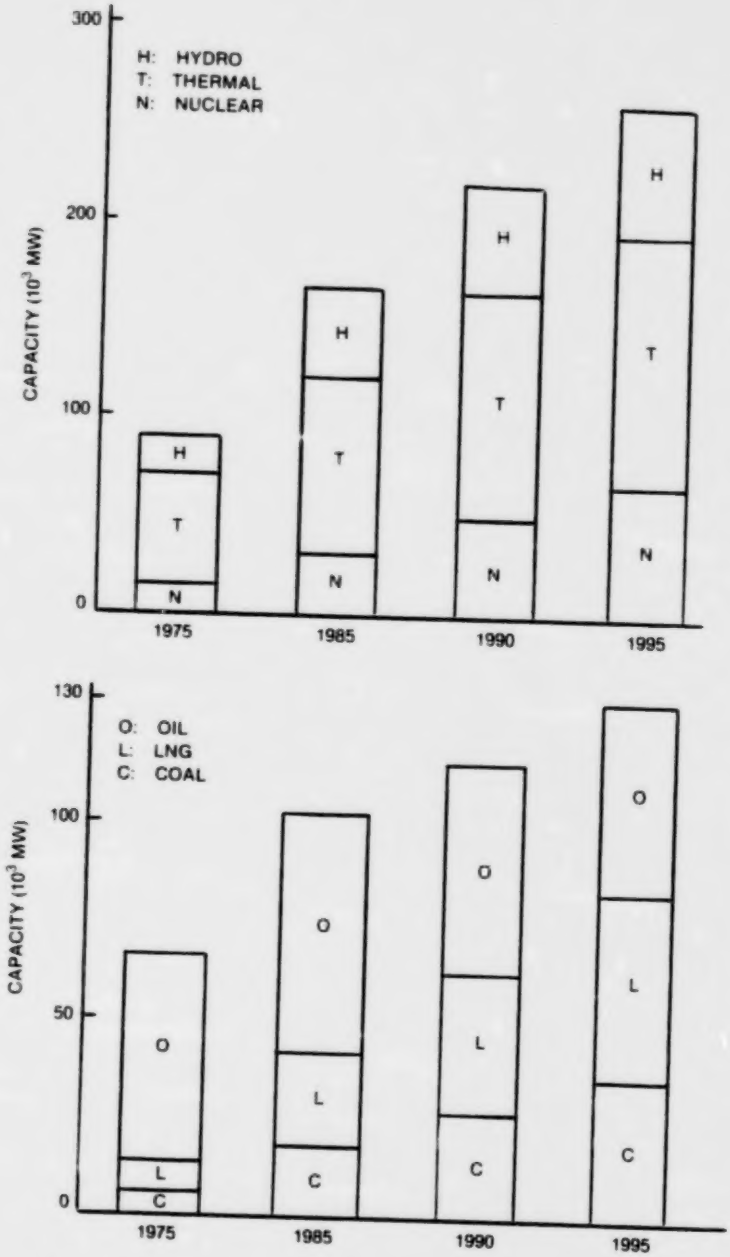


**Figure 1-6** Estimated Increase in Electrical Power Generation Capacity in Japan: 1975-1995.

TABLE 1-5. ESTIMATED DEMAND AND SUPPLY OF FUEL COAL ($10^6$ TON)

| | 1978 | 1985 | 1990 | 1995 |
|---|---|---|---|---|
| **Demand** | | | | |
| Power Generation | 7.7 | 16.5 | 42.0 | 66.0 |
| Cement | 0.5 | 10.0 | 11.0 | 12.0 |
| Others | 2.6 | 8.5 | 13.5 | 15.5 |
| Total | 10.8 | 35.0 | 66.5 | 93.5 |
| **Supply** | | | | |
| Domestic | 9.8 | 13.0 | 13.0 | 13.0 |
| Imported | 1.0 | 22.0 | 53.5 | 80.5 |

large population is concentrated in a small land space. The new boilers will be located in regions relatively far from large cities and industrial districts, where the environmental regulations established by the Central Government are not as stringent. However, in order to construct a large plant, it will still be necessary to reach an agreement with the local government, which necessitates extensive countermeasures for pollution control.

### 1.2.2 Coal-Fired Utility Boilers and $SO_x$ and $NO_x$ Removal

Japanese coal-fired utility boilers larger than 250 MW are shown in Tables 1-6a and b. Virtually all of the boilers in operation, under construction, and being designed, use limestone scrubbing to remove 90-95% of the $SO_2$ while by-producing salable gypsum. The outlet $SO_2$ concentration for these systems ranges from 20 to 150 ppm.

$NO_x$ concentrations in the flue gas from coal-fired boilers have been lowered to 150-300 ppm; the emission standard set by the Central Government is 400 ppm. Further $NO_x$ reduction is desired by local governments. Most of the new boilers will install selective catalytic reduction (SCR) units to remove about 80% of $NO_x$ (3).

TABLE 1-6a. COAL-FIRED BOILERS LARGER THAN 250 MW TO BE COMPLETED BY 1986 IN JAPAN

| Power Company | Power Station | Boiler No. | Capacity MW | Year of Completion | | |
|---|---|---|---|---|---|---|
| | | | | Boiler | FGD | SCR[a] |
| Chugoku Electric | Shin-Onoda | 1 | 500 | 1985 | 1985 | 1985 |
| Chugoku Electric | Shin-Onoda | 2 | 500 | 1986 | 1986 | 1986 |
| EPDC[b] | Isogo | 1 | 265 | 1967 | 1976 | |
| EPDC | Isogo | 2 | 265 | 1969 | 1976 | |
| EPDC | Takasago | 1 | 250 | 1968 | 1975 | |
| EPDC | Takasago | 2 | 250 | 1969 | 1976 | |
| EPDC | Takehara | 1 | 250 | 1967 | 1977 | 1981 |
| EPDC | Takehara | 3 | 700 | 1982 | 1982 | 1982 |
| EPDC | Matsushima | 1 | 500 | 1981 | 1981[c] | |
| EPDC | Matsushima | 2 | 500 | 1981 | 1981[c] | |
| EPDC | Matsuura | 1 | 1000 | 1986 | 1986 | 1986 |
| Hokkaido Electric | Tomato-Atsuma | 1 | 350 | 1980 | 1980[d] | 1980[e] |
| Hokkaido Electric | Tomato-Atsuma | 2 | 600 | 1984 | 1984 | |
| Joban Joint Power | Nakoso | 8 | 600 | 1983[f] | 1983 | 1983 |
| Joban Joint Power | Nakoso | 9 | 600 | 1983[f] | 1983 | 1983 |
| Kyushu Electric | Matsuura | 1 | 700 | 1985 | 1985 | 1985 |
| Sakata Joint Power | Sakata | 1 | 350 | 1984 | 1984 | |
| Sakata Joint Power | Sakata | 2 | 350 | 1985 | 1985 | |
| Tohoku Electric | Hachinohe | 5 | 250 | 1986 | 1986 | 1986 |
| Tokyo Electric | Yokosuka | 1 | 265 | 1984[g] | 1984 | 1984 |
| Tokyo Electric | Yokosuka | 2 | 265 | 1984[g] | 1984 | 1984 |
| Toyama Joint Power | Toyama | 1 | 200[h] | 1984 | 1984 | |
| Toyama Joint Power | Toyama | 2 | 200[h] | 1984 | 1984 | |

(Continued)

TABLE 1-6b. COAL-FIRED BOILERS LARGER THAN 250 MW TO BE COMPLETED BETWEEN 1987 AND 1991 (Continued)

| Power Company | Power Station | Boiler No. | Capacity MW | Year of Completion |
|---|---|---|---|---|
| Chubu Electric | Hekinan | 1 | 700 | 1989 |
| Chubu Electric | Hekinan | 2 | 700 | 1989 |
| Chugoku Electric | Misumi | 1 | 700 | 1989 |
| Chugoku Electric | Misumi | 2 | 700 | 1990 |
| EPDC | Matsuura | 2 | 1000 | 1991 |
| Hokkaido Electric | Tomato-Atsuma | 3 | 600 | 1987 |
| Hokuriku Electric | Tsuruga | 1 | 500 | 1987 |
| Hokuriku Electric | Tsuruga | 2 | 500 | 1987 |
| Kyushu Electric | Matsuura | 2 | 1000 | 1991 |
| Kyushu Electric | Reihoku | 1 | 700 | 1988 |
| Kyushu Electric | Reihoku | 2 | 700 | 1989 |
| Soma Joint Power | Soma | 1 | 1000 | 1989 |
| Soma Joint Power | Soma | 2 | 1000 | 1991 |
| Tohoku Electric | Haramachi | 1 | 1000 | 1988 |
| Tohoku Electric | Haramachi | 2 | 1000 | 1991 |
| Tohoku Electric | Noshiro | 1 | 600 | 1987 |
| Tohoku Electric | Noshiro | 2 | 600 | 1991 |
| Tokyo Electric | Mito | 1 | 1000 | 1989 |
| Tokyo Electric | Mito | 2 | 1000 | 1989 |

[a] Selective catalytic reduction of $NO_x$.
[b] Electric Power Development Co.
[c] Treating three-fourths of gas
[d] Treating one-half of gas
[e] Treating one-fourth of gas
[f] Both coal and oil will be used
[g] Existing boiler using oil; coal-oil mixture will be used from 1984
[h] Existing 250 MW oil-fired boiler will burn coal at 200 MW

As shown in Table 1-7, many old smaller boilers that originally burned coal switched to oil and now are using coal again. Most of those boilers have installed not only FGD but also SCR units, because the desired level of $NO_x$ reduction is difficult to achieve by combustion modification with the older boilers. Chugoku Electric has installed both FGD and SCR on its Shimonoseki No. 1 boiler and Shin-Ube No. 1 - No. 3 boilers, and successfully switched from oil to coal. This success has encouraged other power companies to make similar changes.

TABLE 1-7. EXISTING UTILITY BOILERS ORIGINALLY DESIGNED FOR COAL

| Power Company | Station | Boiler No. | Capacity (MW) | Year of Completion | |
|---|---|---|---|---|---|
| | | | | FGD | SCR |
| Chugoku Electric | Shimonoseki | 1 | 175 | 1979 | 1980 |
| Chugoku Electric | Shin-Ube | 1 | 75 | 1982 | 1982 |
| Chugoku Electric | Shin-Ube | 2 | 75 | 1982 | 1982 |
| Chugoku Electric | Shin-Ube | 3 | 156 | 1982 | 1982 |
| Chugoku Electric | Mizushima | 1 | 125 | 1984 | 1984 |
| Chugoku Electric | Mizushima | 2 | 156 | 1984 | 1984 |
| Kyushu Electric | Omura | 2 | 156 | 1982 | 1982 |
| Kyushu Electric | Minato | 1 | 156 | 1983 | 1983 |
| Tohoku Electric | Sendai | 2 | 175 | 1983 | 1983 |
| Tohoku Electric | Sendai | 3 | 175 | 1983 | 1983 |
| Shikoku Electric | Saijo | 1 | 175 | 1983 | |
| Shikoku Electric | Saijo | 2 | 250 | 1984 | |

Coal-fired industrial boilers are not popular in Japan. There are two in operation (156 MW and 175 MW), both of which have FGD units. Few coal-fired industrial boilers will be installed in the future because of the difficulty involved in obtaining land space for coal storage and in gaining the permission of the local governments.

The FGD plants in operation for coal-fired boilers are described in Sections 3 and 4 of this report.

### 1.2.3 Minor Components of Coal and Fly Ash

Table 1-8 shows concentrations of minor elements in coal compared with those in oil and the earth's crust. The elements present in coal in concentrations more than ten times larger than in the earth's crust are boron (B), cadmium (Cd), zinc (Zn), and chlorine (Cl). Among those elements, cadmium can be toxic but may not cause problems because of the low concentration in which it is found. The elements present in coal in concentrations less than one-tenth that in earth's crust are chromium (Cr), fluorine (F), manganese (Mn), and Thorium (Th). The concentrations of many other elements found in coal are similar to those in the earth's crust. Compared with oil, coal is rich in most elements except molybdenum (Mo) and vanadium (V). When coal is combusted, most of those elements (except chlorine, fluorine, and mercury) are present in the ash. Fly ash from pulverized coal consists of fine particles mostly ranging in size from 1 to 50 microns. Over 99.8% of the fly ash, chlorine, and fluorine, as well as most of the mercury and other metals in the flue gas can be removed with the combined use of electrostatic precipitators (ESPs) and wet flue gas desulfurization systems. Dust removal using baghouses also has been studied in Japan but will probably not become popular for utility boilers because of the large pressure drop and high operation cost associated with it.

In 1978 about 2 million tons of fly ash were produced in Japan. Over 70% of the ash was used for landfill while the rest was used for cement and other materials. Examples of test results on the leaching of ash components are given in Tables 1-9 and 1-10. The concentrations of the dissolved components are low enough to meet applicable regulations.

The most significant problem associated with increased coal usage in Japan is the disposal of fly ash, because land disposal space is limited and landfilling on the sea coast has been restricted to protect the environment. New uses for ash such as asphalt filler and raw material for aggregate have been studied. A small amount of the fly ash also can be used as a soil conditioner to supply micronutrients such as boron and zinc to plants.

TABLE 1-8. CONCENTRATION OF ELEMENTS IN COAL, OIL, AND THE EARTH'S CRUST (ppm)

| Element | Coal[a] |  |  | Oil[b] | Earth | A/B |
|---|---|---|---|---|---|---|
|  | Average (A) | Min. | Max. | (ppm) | Crust (B)[c] |  |
| As | 14.02 | 0.53 | 93.00 | 0.26 | 2 | 7 |
| B | 102.21 | 5.00 | 224.00 | 0.002 | 3 | 34 |
| Be | 1.61 | 0.20 | 4.00 | <0.003 | 2 | 1 |
| Cd | 2.52 | 0.10 | 65.00 | 0.03 | 0.15 | 17 |
| Co | 9.57 | 1.00 | 43.00 | 0.05 | <20 | 1 |
| Cr | 13.75 | 4.00 | 54.00 | 0.05 | 100 | 0.1 |
| Cu | 15.16 | 5.00 | 61.00 | 1.3 | 55 | 0.3 |
| F | 60.94 | 25.00 | 143.00 | — | 700 | 0.1 |
| Hg | 0.20 | 0.02 | 1.60 | 0.05 | 0.08 | 2.5 |
| Mn | 49.4 | 6.00 | 181.00 | 0.6 | 1000 | 0.05 |
| Mo | 7.54 | 1.00 | 30.00 | 80.00 | 10 | 0.7 |
| Ni | 21.07 | 3.00 | 80.00 | 10 | <35 | 1 |
| Pb | 34.78 | 4.00 | 218.00 | — | 15 | 2 |
| Sb | 1.26 | 0.20 | 8.90 | 0.05 | 0.1~0.2 | 10 |
| Se | 2.08 | 0.45 | 7.70 | 0.03 | 0.09 | 23 |
| V | 32.71 | 11.00 | 78.00 | 50 | 120 | 0.3 |
| Zn | 272 | 6.00 | 5350 | 30 | 40 | 63 |
| V | 1.6 | 0.5 | 4.5 | 0.015 | 3-4 | 0.5 |
| Th | 2.01 | 1.2 | 3.3 | — | 10-15 | 0.1 |
| Cl | 1400 | 100 | 5400 | 39 | 150 | 9.3 |

[a] N.Y. Lim, "Trace Elements from Coal Combustion Atmospheric Emissions," IEA-Coal Research, London ICTIS/TRO 5 (1979).

[b] Environmental Chemistry (1979)

[c] Ahrens-Tayler (1961), etc.

TABLE 1-9. EXAMPLE OF DISSOLUTION TEST OF HAZARDOUS COMPONENTS FROM FLY ASH (mg/1) (Environment Agency, Japan)

| Component | Type of Coal |  | Regulations |  |
|---|---|---|---|---|
|  | Domestic | Imported | Landfill | Ocean Dumping |
| Hg (organic) | ND[a] | ND[a] | ND[a] | ND[a] |
| Hg (total) | ND[a] | ND[a] | <0.005 | <0.005 |
| Cd | <0.018 | ND[a] | <0.3 | <0.1 |
| Pb | <0.02 | ND[a] | <3 | <1 |
| P (organic) | ND[a] | ND[a] | <1 | <1 |
| $Cr^{6+}$ | <0.20 | <0.10 | <1.5 | <0.5 |
| As | <0.012 | <0.38 | <1.5 | <0.5 |
| CN | <0.01 | ND[a] | <1 | <1 |

[a] Not detected

TABLE 1-10. SAMPLE FLY ASH COMPONENT DISSOLUTION TESTS (mg/1) (Environment Agency, Japan)

| Component | Type of Coal |  | Emission Standards |
|---|---|---|---|
|  | Domestic | Imported |  |
| COD | 0.7 | 1.6 | 120 |
| BOD | 1.6 | --- | 120 |
| Phenol | ND[a] | ND[a] | 5 |
| Cu | 0.18 | ND[a] | 3 |
| Zn | 0.08 | ND[a] | 5 |
| Mn | 0.02 | ND[a] | 10 |
| Cr (total) | ND[a] | 0.18 | 2 |
| F | 0.11 | 1.02 | 15 |
| PH | 12.5 | 10.6 | 5.8 - 8.6 |

[a] Not detected

### 1.2.4 New Uses for Coal

In an attempt to solve the coal handling problem, Japanese researchers have extensively studied the use of a coal-oil fuel mixture (COM); it is planned for use in some boilers including those at Tokyo Electric's Yokosuka Station (Table 1-6). The major drawback associated with COM is that more than half of its energy is derived from oil. A coal-water slurry (70-75% coal) also has been tested.

Fluidized bed combustion (FBC), gasification, and liquefaction of coal, are all being studied, although not as extensively as in the U.S. FBC research is described in Section 1.3. The major problem with FBC in Japan is the difficulty encountered in disposal of fly ash containing lime, calcium sulfate, and sulfite.

Coal gasification tests are described in Section 1.4. Coal gasification as well as liquefaction may not be suitable in Japan which must import coal; a considerable portion of the energy of coal is consumed by the gasification or liquefaction process. Importing large amounts of LNG and using it for power generation decreases the need for coal gasification.

Tests on coal liquefaction have also been conducted in Japan. To date, the largest is the Brown Coal Liquefaction Project, jointly sponsored by a group of Japanese companies and the Australian government to produce liquid fuel. Liquefaction tests have also been carried out for production of binder for non-coking coal for coke production. Although liquefaction may become more important in future, it would make more sense for a plant to be constructed abroad near the coal mine and the product subsequently exported to Japan.

Several cement kilns in Japan have begun using coal to replace oil. Coal is suitable for cement kilns because most of the $SO_x$ in flue gas is absorbed by the large amount of lime in the kiln. In addition, the fly ash from coal reacts with lime to form cement. The high $NO_x$ concentration in the flue gas (usually from 400 - 800 ppm) presents a considerable problem in Japan. However, by using a modified suspension preheater, the $NO_x$ concentration can be lowered to 200 ppm or below.

Steel producers have been testing the injection of pulverized coal or a coal-oil mixture as a substitute for oil in blast furnaces. The use of coal or coal-oil mixtures does not appreciably increase environmental problems because most of the $SO_x$ and ash are trapped in the furnace and little $NO_x$ is formed due to the reducing nature of the furnace gas. Since November 1981 Sumitomo Metal has been using 20 t/hr of a coal-oil mixture for the No. 3 blast furnace at its Kashima Works.

## 1.3 FLUIDIZED BED COMBUSTION (4)

### 1.3.1 Introduction

Tests on atmospheric fluidized bed combustion (AFBC) of coal have been carried out since 1978 under the leadership of the Ministry of International Trade and Industry (MITI). Following small scale tests, construction of a pilot plant with a generating capacity of 20 tons/hr steam (5 MW) was completed in 1980 (see Figure 1-7). Ongoing tests at the plant are being conducted by the Coal Mining Research Center jointly with Electric Power Development Co. (EPDC), Kawasaki Heavy Industries (KHI) and Babcock Hitachi K.K. (BHK). The pilot plant is located at EPDC's Wakamatsu Station. The total budget for the construction and operation of the pilot plant is 2 billion yen. Two-thirds of the project funding came from MITI, with the rest provided by EPDC, KHI, and BHK.

In 1980, Japan joined the International Energy Agency's (IEA's) international cooperative research project on atmospheric fluidized bed combustion. Test results from the Wakamasu pilot plant will be used in the IEA project.

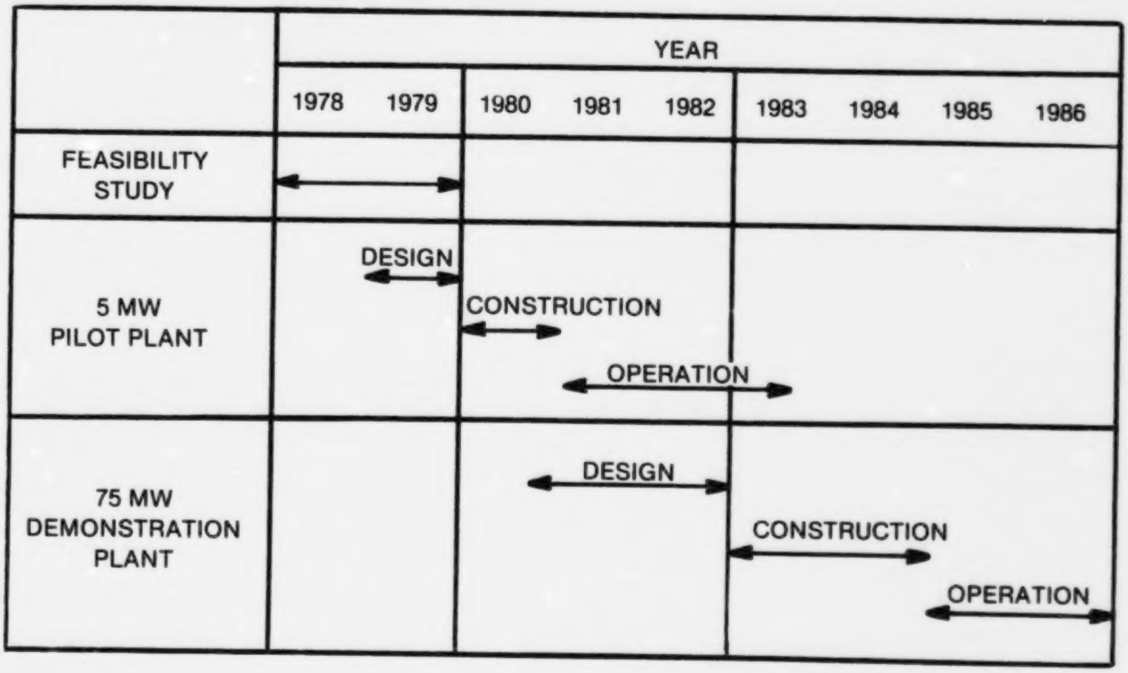


**Figure 1-7** **MITI's FBC Development Plant.**

### 1.3.2 Process Description

The design data for the Wakamatsu FBC pilot plant are shown in Table 1-11.

TABLE 1-11. DESIGN DATA FOR THE WAKAMATSU ATMOSPHERIC FBC PILOT PLANT

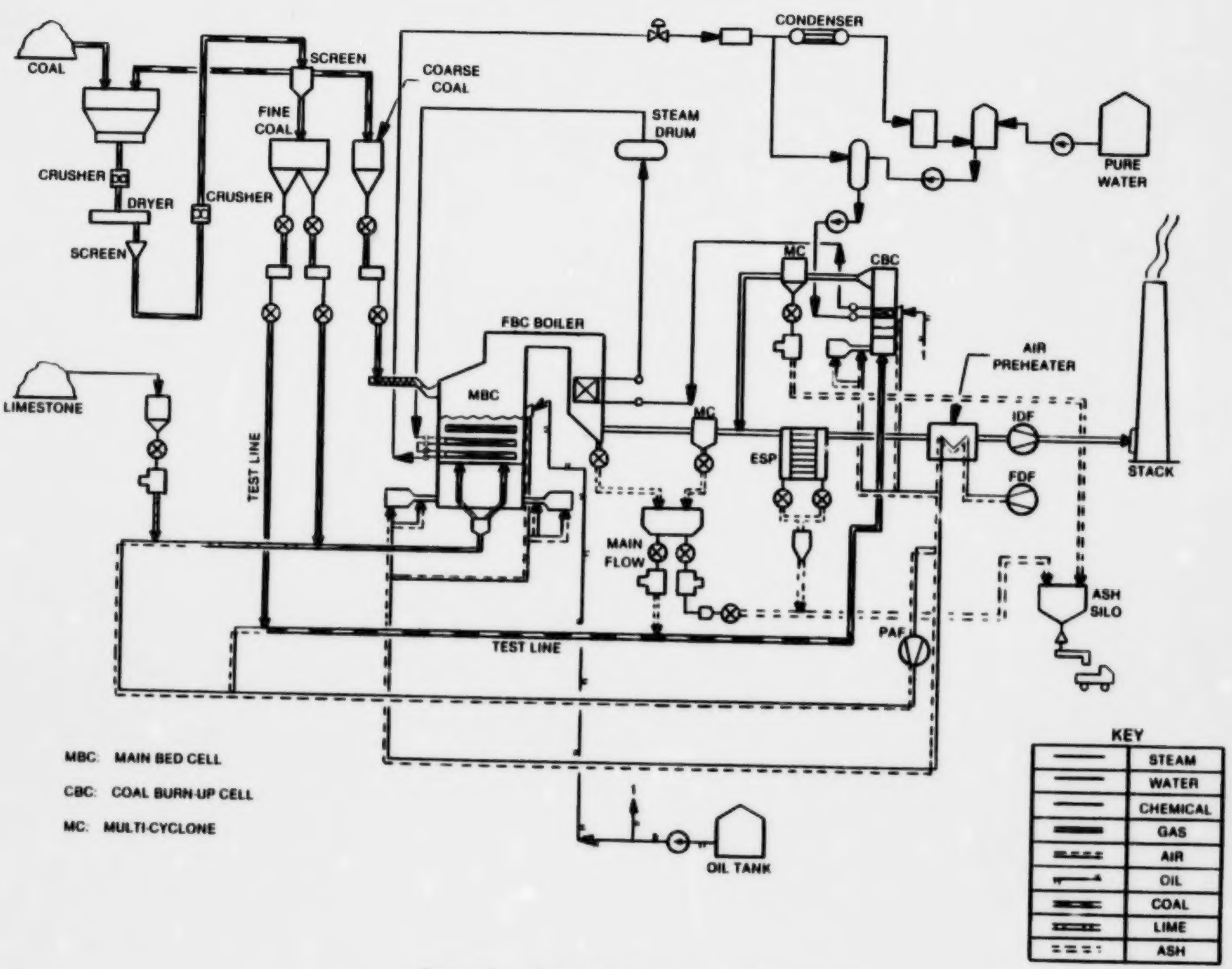
| | |
|---|---|
| Steam generation capacity, t/hr | 20 |
| Steam pressure at superheater outlet, kg/cm²G | 60 |
| Steam temperature at superheater outlet, °C | 540 |
| Feed water temperature at economizer inlet, °C | 133 |
| Boiler efficiency, % | 87.37 |
| Coal consumption, kg/hr | 2,450 (wet) |
| Heat value, kcal/kg | 7,100 (dry)<br>6,603 (wet) |
| Flue gas, Nm³/hr | 20,293 (wet) |
| $SO_x$ removal efficiency (target), % | 95 by limestone or synthetic absorbent |
| $NO_x$ concentration (target), ppm | 60 |

Figure 1-8 shows a flowsheet of the pilot plant. The combustion system consists of a main bed cell (MBC) and a coal burn-up cell (CBC). The latter is used when the combustion of the MBC is not sufficient and yields a carbon-rich ash. MBC and CBC test conditions are shown in Table 1-12.

TABLE 1-12. TEST CONDITIONS OF MBC AND CBC

| | MBC | CBC |
|---|---|---|
| Total air ratio | 1.1 - 1.25 | 1.2 - 1.4 |
| Bed temperature, °C | 770 - 900 | 900 - 1,000 |
| Height of fluidized zone, m | 1.2 - 1.6 | 1.2 - 1.6 |
| Fluidizing medium | Limestone | Silica sand |
| $SO_x$ absorbent | Limestone | |
| Coal size, mm | Under 5 - Under 12 | |
| $CaO/SO_x$ mole ratio | Below 5 | |
| Air ratio in bed | 0.85 - 1.25 | 0.9 - 1.4 |
| Heat load, kcal/m³ hr | $1.5 - 2.7 \times 10^5$ | |

Both MBC and CBC have over-fire air ports. This enables combustion in the bed to be carried out in a reducing atmosphere, thereby reducing $NO_x$.


COAL
CRUSHER
DRYER
SCREEN
CRUSHER
SCREEN
FINE COAL
COARSE COAL
STEAM DRUM
CONDENSER
PURE WATER
LIMESTONE
TEST LINE
FBC BOILER
MBC
MC
CBC
MC
ESP
MAIN FLOW
TEST LINE
AIR PREHEATER
IDF
FDF
STACK
PAF
ASH SILO
OIL TANK
KEY
STEAM
WATER
CHEMICAL
GAS
AIR
OIL
COAL
LIME
ASH

MBC: MAIN BED CELL

CBC: COAL BURN-UP CELL

MC: MULTI-CYCLONE

**Figure 1-8** Flowsheet of Wakamasu AFBC Pilot Plant.

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As shown in Table 1-13, pilot plant testing began in May 1981 with domestic low-sulfur coal. Preliminary results indicated that with combustion at 805°C, the $NO_x$ level was 189 ppm without staged combustion and 64 ppm with staged combustion. The $SO_x$ level was 0.12 $Nm^3/hr$ (about 60 ppm). Since November 1981 medium-sulfur coal has been used to test for $SO_x$ removal. Later, a synthetic $SO_x$ absorbent will be used which may be separated from ash, regenerated and recycled for $SO_x$ removal.

TABLE 1-13. PROPERTIES AND COMPOSITION OF COAL USED AT THE WAKAMASU AFBC PILOT PLANT

| | |
|---|---|
| Heat value, kcal/kg | 6840 |
| Ash, % | 13.2 |
| Volatile matter, % | 44.5 |
| Fixed carbon, % | 40.1 |
| Total carbon, % | 65.7 |
| Nitrogen, % | 0.7 |
| Sulfur (combustible), % | 0.1 |
| Sulfur (noncombustible), % | 0.1 |

### 1.3.3 Evaluation

Coal combustion in an atmospheric fluidized bed was successfully achieved in the pilot plant. The resulting low $NO_x$ concentration--64 ppm with staged combustion--is attractive. Various coals will be tested in the future.

The major deterrent to FBC in Japan is ash disposal. When over 3 mols $CaCO_3$ are used per mol of $SO_x$, 90% of the $SO_x$ is removed, but it is difficult to dispose of the ash which contains considerable amounts of lime, calcium sulfate, and sulfite. Although it may be possible to use an $SO_x$ absorbent which can be separated from fly ash and regenerated for reuse, the process may have technical and economic disadvantages.

In Japan, a small commercial FBC unit (20 ton/hr) has been operated which burns low-calorie coal (3,000 kcal/kg) recovered from the wash water of low-sulfur coal. In this unit silica sand is used as the fluidizing medium without desulfurization (5). FBC may be appropriate for such applications.

## 1.4 COAL GASIFICATION (LOW CALORIE GAS) (6)

### 1.4.1 Introduction

The Coal Mining Research Center has conducted coal gasification tests sponsored by MITI to produce low calorie gas for electrical power generation. The test development plan is shown in Figure 1-9. Since 1975 a pilot plant which uses wet desulfurization and has a capacity of gasifying 5 tons/day of coal has been operated at Yubari, Hokkaido. A larger plant with a capacity of gasifying 40 tons/day coal was completed at the end of 1980. A dry desulfurization unit using iron oxides is now under construction.

If the test results at these sites are promising, a 1,000 ton/day demonstration plant may be constructed in 1985.

### 1.4.2 Process Description

The gasifier tested by the Coal Mining Research Center consists of a two-stage fluidized bed furnace as shown in Figures 1-10 and 1-11. Pulverized coal with an average size of 1 - 1.5 mm, and a moisture content of less than 3%, is fed into the upper furnace and burned at a temperature above 800°C. In the furnace, volatile matter is gasified and tar is thermally decomposed to form char. Fine particles of char (smaller than 0.6 mm) leave the reactor with the flue gas and are trapped by a cyclone, while larger char particles are discharged from the middle portion of the upper furnace. The char is fed into the bottom furnace and gasified with air and steam at 650-750°C. The gas formed in the bottom furnace is mixed with the gas formed in the upper furnace to yield a crude gas with a heat value of 1,200 kcal/Nm$^3$.

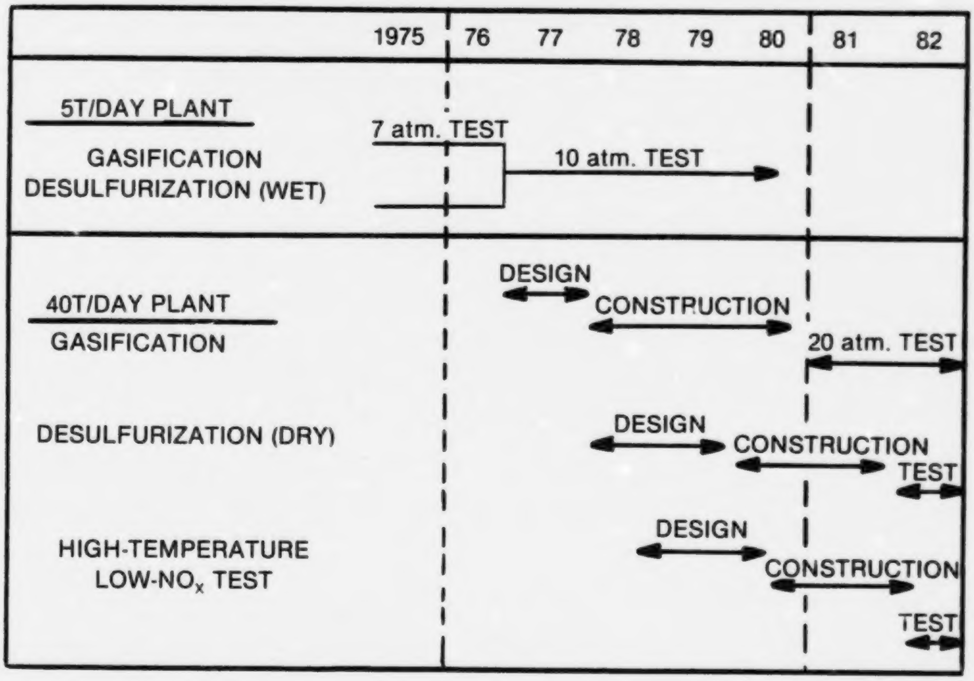

**Figure 1-9** MITI's Development Plan for Coal Gasification.

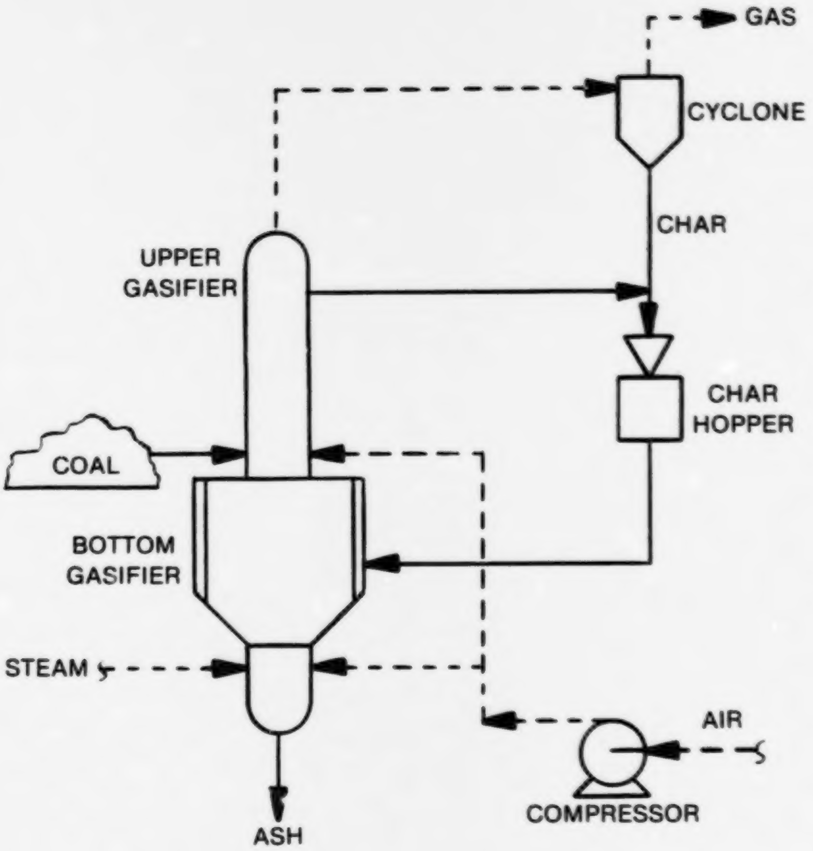
GAS
CYCLONE
CHAR
UPPER GASIFIER
CHAR HOPPER
COAL
BOTTOM GASIFIER
STEAM
AIR
COMPRESSOR
ASH

**Figure 1-10** **Coal Mining Research Center's Gasification System**

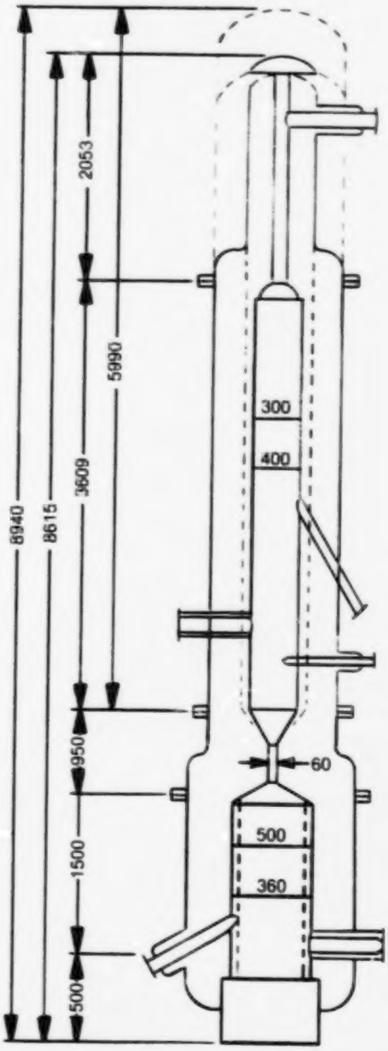
8940
8615
5990
2053
3609
950
1500
500
300
400
60
500
360

**Figure 1-11 Coal Mining Research Center's Gasifier.***

*Dimensions shown in mm. Solid line indicates original shape; dotted line indicates the shape after modification.

Next, the crude gas is passed through two cyclones in series, washed with water, and subjected to $H_2S$ removal by the Benfield process using potassium carbonate scrubbing. The absorbed $H_2S$ is stripped from the liquor with steam and is incinerated; $H_2S$ also may be used for elemental sulfur production at a commercial unit.

Two domestic low-sulfur coals have been used for the tests (Table 1-14). The Taiheiyo coal is non-coking, while the Horonai coal has an insignificant coking property.

TABLE 1-14. COMPOSITION OF COALS USED DURING GASIFIER TESTING

| | Taiheiyo | Horonai |
|---|---|---|
| Moisture, % | 6.0~6.4 | 3.4~3.5 |
| Ash, % | 15.5~17.5 | 12.3~13.5 |
| Volatile Matter, % | 39.0~40.5 | 40.1~41.8 |
| Fixed Carbon, % | 36.5~38.0 | 41.3~43.5 |
| Total Carbon, % | 62.5~64.5 | 68.6~70.6 |
| H, % | 5.1~5.3 | 5.3 |
| N, % | 1.0~1.2 | 1.1 |
| O, % | 12.0~12.7 | 10.0~11.0 |
| S, % | 0.27~0.33 | 0.26~0.27 |
| Heat Value, kcal/kg | 5920~6260 | 6750~6850 |

## 1.4.3 Results of Gasification Performance Tests

One of the major problems revealed during testing of the gasification process was clinker formation in the bottom furnace. Combustion at a low temperature prevents this formation but lowers the gasification efficiency. The clinker formation problem has been solved in the following way: 1) using a suitable gas velocity (about 1 m/sec), suitable coal size (average 1.5 mm) and a special air-sparging plate (4); and 2) adjusting the temperature to 650-750°C. The temperature can be adjusted by changing the steam/air ratio as shown in Figure 1-12. The relationship between the steam/air ratio and the gas composition is shown in Figure 1-13.

During testing it was demonstrated that when the temperature of the upper furnace was lower than that of the bottom furnace, a considerable

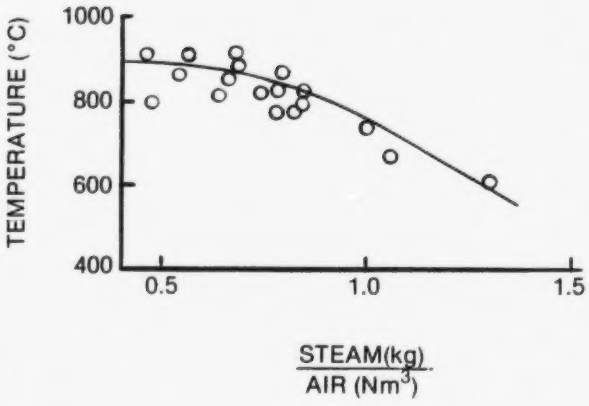

Figure 1-12 Steam/Air Ratio and Temperature of the Bottom Furnace during Gasification Testing.

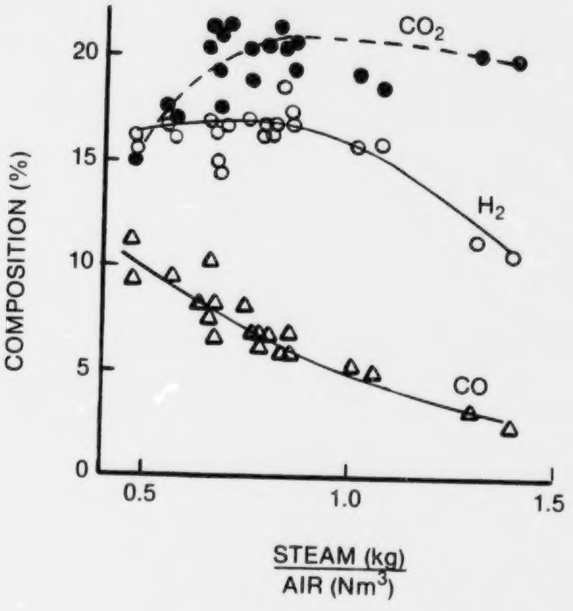

Figure 1-13 Steam/Air Ratio and Gas Composition of the Bottom Furnace during Gasification Testing.

number of tar deposits formed inside the vessels and caused problems. To minimize tar deposits, air was introduced into the upper furnace in order to raise the temperature above 800°C.

Two examples of the carbon balance with Taiheiyo coal are shown in Figure 1-14 and Tables 1-15 and 1-16. When the temperature of the upper furnace was 704°C, only 78.24% of the carbon in the coal was recovered as gas; considerable portions of the carbon went into char and tar. At 850°C the carbon recovery increased to 87.02%.

TABLE 1-15. CARBON BALANCE (UPPER FURNACE 704°C)

| | 1 | 2 | 3 | 4 | 5 | Total | 6 |
|---|---|---|---|---|---|---|---|
| Recovery from 100 kg Coal | 5.64<br>(kg) | 0<br>(kg) | 4.21<br>(kg) | 14.55<br>(kg) | ---<br>--- | ---<br>--- | 371.4<br>(Nm$^3$) |
| Carbon Content, % | 8.22 | --- | 42.34 | 36.39 | --- | --- | 13.33 |
| Amount of Carbon, kg | 0.46 | --- | 1.78 | 5.29 | 6.19 | 13.72 | 49.45 |
| Yield of Carbon, % | 0.73 | --- | 2.82 | 8.38 | 9.82 | 21.76 | 78.24 |

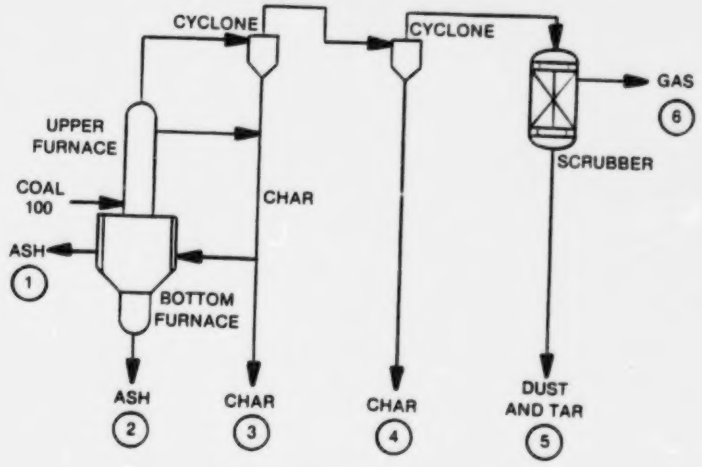
CYCLONE
CYCLONE
GAS
⑥
UPPER FURNACE
SCRUBBER
COAL 100
CHAR
ASH
①
BOTTOM FURNACE
ASH
②
CHAR
③
CHAR
④
DUST AND TAR
⑤

Figure 1-14 Carbon Balance with Taiheiyo Coal.

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TABLE 1-16. CARBON BALANCE (UPPER FURNACE 850°C)

|  | 1 | 2 | 3 | 4 | 5 | Total | 6 |
|---|---|---|---|---|---|---|---|
| Recovery from 100 kg Coal | 2.34 (kg) | 1.28 (kg) | 0 (kg) | 14.30 (kg) | — | — | 400.7 ($Nm^3$) |
| Carbon Content, % | 12.59 | 2.46 | — | 24.81 | — | — | 14.24 |
| Amount of Carbon, kg | 0.29 | 0.03 | — | 3.55 | 4.64 | 8.51 | 57.06 |
| Yield of Carbon, % | 0.45 | 0.05 | — | 5.41 | 7.07 | 12.98 | 87.02 |

Horonai coal was also gasified without any problems caused by its slight coking nature but its gasification efficiency tended to be slightly lower than that of Taiheiyo coal.

### 1.4.4 Evaluation

The gasification tests conducted by the Coal Mining Research Center resulted in the solution of various problems associated with the process. However, using the synthetic gas for power generation by combined cycle also may have problems. Combined cycle power generation using kerosene has been commercialized successfully in Japan using selective catalytic reduction of $NO_x$ (3). A similar system using LNG is expected to be commercialized in the near future. Compared with these systems, the use of gasified coal to generate power has had many more difficulties due to particulates and $H_2S$ in the gas. Removal of these pollutants using a wet process causes a considerable energy loss making an efficient dry removal process preferable.

Since Japan plans to import a large amount of LNG to use primarily for power generation (Figure 1-6), power generation by coal gasification may not be commercialized unless the technical and environmental problems associated with it are solved.

# SECTION 2

# GENERAL ASPECTS OF FLUE GAS DESULFURIZATION (FGD)

## 2.1 MAJOR FGD PROCESSES AND BY-PRODUCTS

The number of FGD systems commercially available in Japan has increased markedly since 1968. About 1200 systems, primarily controlling oil-fired boiler emissions, are presently in operation. Table 2-1 lists the approximate number and capacities of these units by process.

TABLE 2-1. APPROXIMATE NUMBER AND CAPACITY OF JAPANESE FGD UNITS PRESENTLY IN OPERATION

| Process | Number | Capacity (1,000 $Nm^3/hr$) |
|---|---|---|
| $Na_2SO_3$ $Na_2SO_4$ byproduct | 1,000 | 50,000 |
| Wet lime/limestone | 110 | 55,000 |
| Indirect lime/limestone | 50 | 15,000 |
| $H_2SO_4$, S, $(NH_4)_2SO_4$ byproduct | 30 | 12,000 |

Sodium scrubbing which produces sodium sulfite or sulfate byproducts is the simplest FGD system. The sulfite can be sold to paper mills and the sulfate can be used for glass production. Over 700 sodium scrubbing units with capacities of 3,000 - 300,000 $Nm^3/hr$ (1-100 MW equivalent) were constructed between 1968 and 1974. This construction resulted in an oversupply of sulfite and sulfate (Figure 2-1). Therefore, only larger sodium-scrubbing units presently practice byproduct recovery while many smaller units purge the sulfite or sulfate in wastewater.

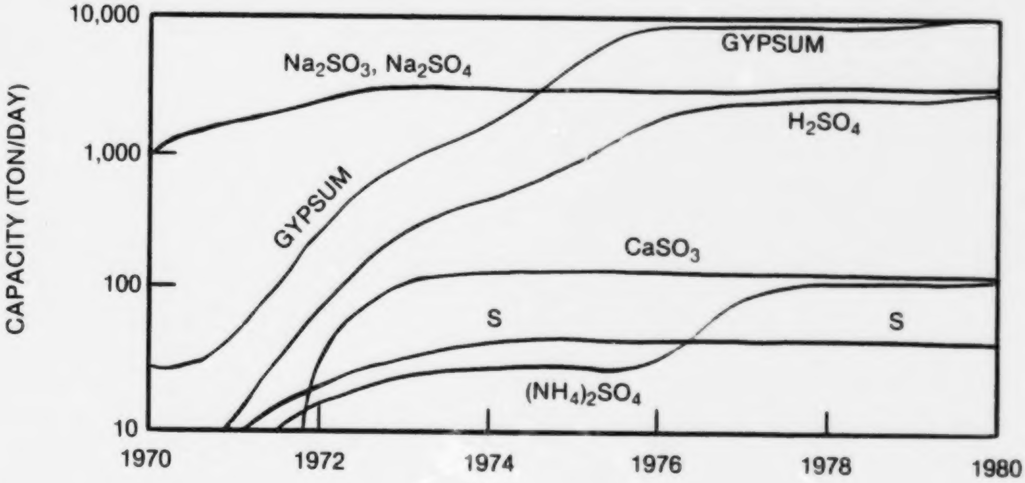


**Figure 2-1** By-product Production Capacity of FGD Systems: 1970-1980.

Most of the utility boilers use wet lime/limestone scrubbing which produces salable gypsum because the cost is relatively low and gypsum can be used for cement setting retarder and wallboard. In 1975, gypsum was in oversupply and had no sale value. Since 1976, demand and supply have balanced and gypsum prices have recovered (Figures 2-2 and 2-9).

There are presently about 50 units using an indirect lime/limestone FGD process. With this process, $SO_2$ is absorbed in a liquor which contains sodium sulfite, aluminum sulfate or dilute sulfuric acid and ferric sulfate. The liquor is treated with lime or limestone to precipitate and separate calcium sulfite or gypsum. The indirect process is used mainly for medium-sized gas sources with a capacity of 50,000 - 500,000 $Nm^3$/hr since scaling problems for units of this size can be prevented easily. However, it is generally more expensive than the lime/limestone process.

There are about 20 FGD units presently producing a sulfuric acid byproduct in Japan. Most of them use the Wellman-Lord process to recover concentrated $SO_2$ for use in acid production. However, several units at oil refineries use the concentrated $SO_2$ to produce elemental sulfur by the Claus process.

There are two large ammonia scrubbing units using ammonia in coke oven gas to recover $SO_2$ from boilers for ammonium sulfate production. These recovery processes are not used extensively at power plants because sulfuric acid or sulfur production processes are more costly than limestone-gypsum processes, and ammonium sulfate is currently in oversupply.

Almost all Japanese FGD units use wet processes. Dry processes were studied in the early stages of FGD development, but most were not used because early investigation showed them to have low $SO_2$ removal efficiencies and high costs. Presently, there are only 4 dry process units in operation--3 of them use carbon adsorption and one uses copper oxide absorption. A few units use a semi-dry process in which an alkaline slurry is sprayed and a powdery sodium sulfite is recovered. A semi-dry process developed in the U.S. involves spraying lime slurry to obtain a powdery calcium sulfite. This method probably will

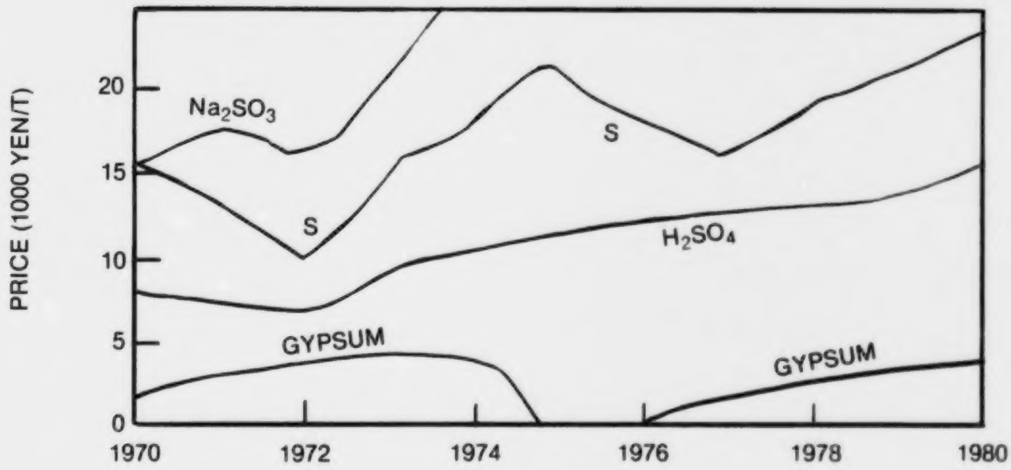


**Figure 2-2** **Price of FGD By-products.**

not be used in Japan because of difficulties with byproduct disposal and with attaining a high $SO_2$ removal efficiency.

## 2.2 FGD PROCESSES AND COAL PLANTS

FGD processes for coal-fired units are listed in Table 2-2. Most of them are wet processes using lime or limestone and one process uses red mud. There are presently three dry processes which have been tested for coal-fired applications using activated carbon or electron beams.

TABLE 2-2. FGD PROCESSES FOR COAL-FIRED UNITS

| FGD Process Developer | Reagent | By-Product |
|---|---|---|
| Mitsui Miike Machinery Co. (MMMC)[a] | $CaO/CaCO_3$ | Sludge/gypsum |
| Ishikawajima-Harima Heavy Industries (IHI)[a] | $CaCO_3$ | Gypsum |
| Babcock Hitachi K. K. (BHK)[a] | $CaCO_3$ | Gypsum |
| Mitsubishi Heavy Industries (MHI)[a] | $CaCO_3$ | Gypsum |
| Chiyoda Chemical Engineering and Construction (Chiyoda)[b] | $CaCO_3$ | Gypsum |
| Kawasaki Heavy Industries (KHI)[a] | MgO and CaO | Gypsum |
| Dowa Mining (Dowa)[b] | Aluminum sulfate and $CaCO_3$ | Gypsum |
| Kobe Steel[b] | $CaCl_2$ and CaO | Gypsum |
| Kureha Chemical Industries (Kureha)[b] | $CH_3COONa$ and CaO | Gypsum |
| Sumitomo Aluminum[b] | Red Mud | Sludge |
| Federation of Electric Power Co.[c] | Activated Carbon | Sulfur |
| Electric Power Development Co. (EPDC) and Sumitomo Heavy Industries (SHI)[d] | Activated Carbon, $NH_3$ | Sulfur |
| Mitsui Mining[d] | Activated Carbon, $NH_3$ | Sulfuric Acid |
| Ebara Corp.[d] | $NH_3$ (Electron Beam) | Ammonium Nitrate sulfate |

[a] Commercial
[b] Tested with coal. Commercial operation with other gases.
[c] Tested with coal.
[d] Tested with coal for simultaneous $SO_x$ and $NO_x$ removal.

Table 2-3 lists commercial FGD units for coal-fired boilers. Mitsui Aluminum's 2 units are for industrial boilers while the rest are for utility boilers. All of the units use wet lime/limestone scrubbing. Mitsui Aluminum's No. 1 unit at Omuta, constructed in 1972, was the first Japanese commercial coal-fired boiler FGD unit and used carbide lime to by-produce a throw-away calcium sulfite sludge. All of the other units produce a salable gypsum byproduct, since land space for throw-away systems is not available.

FGD processes using gypsum byproduct recovery are suitable for coal-fired boiler applications even though the gypsum contains a considerable amount of fly ash from the coal. Since the boilers have an electrostatic precipitator (ESP) upstream of the FGD unit, the byproduct gypsum usually contains less than 5% fly ash with 7-10% moisture after being centrifuged. The Isogo station, however, uses low-sulfur coal for which the conventional cold-side ESP is not as efficient. Therefore, the gypsum contains about 20% fly ash and 15% moisture but is still useful.

Commercial FGD units and their operational problems are described in this Section and in Sections 3 and 4. FGD processes which will be used commercially or tested for coal-fired applications are described in Section 5; Section 6 describes simultaneous $SO_x$ and $NO_x$ removal processes.

## 2.3 COMMERCIAL FGD SYSTEMS FOR COAL-FIRED BOILERS

### 2.3.1 Types of Lime/Limestone FGD Systems

Wet lime/limestone FGD processes in commercial use for coal-fired boilers in Japan are classified into 4 types as shown in Figure 2-3 and Table 2-3. Type I is the throw-away sludge system commonly found in the U.S. Only one unit in Japan, the Mitsui Miike Machinery Co. unit at Omuta, employs a throw-away sludge system. Other units use limestone and oxidize calcium sulfite to gypsum using air.

## TABLE 2-3. COMMERCIAL FGD UNITS FOR COAL-FIRED BOILERS IN JAPAN

| Owner | Plant Site | Boiler No. | MW | N/R[a] | FGD Constructor | Year of Completion | Type See Figure 2-3 |
|---|---|---|---|---|---|---|---|
| Mitsui Aluminum | Omuta | 1 | 156 | R | MMMC[b] | 1972 | I |
| Mitsui Aluminum | Omuta | 2 | 175 | N | MMMC | 1975 | II |
| EPDC | Takasago | 1 | 250 | R | MMMC | 1975 | II |
| EPDC | Takasago | 2 | 250 | R | MMMC | 1976 | II |
| EPDC | Isogo | 1 | 265 | R | IHI[c] | 1976 | III |
| EPDC | Isogo | 2 | 265 | R | IHI | 1976 | III |
| EPDC | Takehara | 1 | 250 | R | BHK[d] | 1977 | III[g] |
| EPDC | Takehara | 3 | 700 | N | IHI | 1982 | III[g] |
| EPDC | Matsushima | 1 | 500x3/4 | N | IHI | 1981 | IV |
| EPDC | Matsushima | 2 | 500x3/4 | N | BHK | 1981 | IV |
| Chugoku Electric | Shimonoseki | 1 | 175 | R | MHI[e] | 1979 | IV[g] |
| Chugoku Electric | Shin-Ube | 1 | 75 | R | MHI | 1982 | IV[g] |
| Chugoku Electric | Shin-Ube | 2 | 75 | R | MHI | 1982 | IV[g] |
| Chugoku Electric | Shin-Ube | 3 | 156 | R | MHI | 1982 | IV[g] |
| Chugoku Electric | Mizushima | 1 | 125 | R | BHK | 1983 | IV[g] |
| Chugoku Electric | Mizushima | 2 | 156 | R | BHK | 1983 | IV[g] |
| Hokkaido Electric | Tomato-Atsuma | 1 | 350x1/2 | N | BHK | 1980 | IV[g] |
| Hokkaido Electric | Tomato-Atsuma | 2 | 600 | N | MHI | 1984 | IV |
| Kyushu Electric | Omura | 2 | 156 | R | MHI | 1982 | IV |
| Kyushu Electric | Minato | 1 | 156 | R | MMMC | 1983 | III |
| Joban Joint | Nakoso | 8 | 600 | N | MHI | 1983 | IV[g] |
| Joban Joint | Nakoso | 9 | 600 | N | MHI | 1983 | IV[g] |
| Tohoku Electric | Sendai | 2 | 175 | R | BHK | 1983 | IV |
| Tohoku Electric | Sendai | 3 | 175 | R | BHK | 1983 | IV |
| Tokyo Electric | Yokosuka | 1 | 265 | R | MHI | 1984 | IV[g] |
| Tokyo Electric | Yokosuka | 2 | 265 | R | MHI | 1984 | IV[g] |
| Shikoku Electric | Saijo | 1 | 175 | R | KHI[f] | 1983 | IV |
| Shikoku Electric | Saijo | 2 | 250 | R | KHI | 1984 | IV |

[a] New or retrofit

[b] Mitsui Miike Machinery Co.

[c] Ishikawajima-Harima Heavy Industries

[d] Babcock Hitachi K. K.

[e] Mitsubishi Heavy Industries

[f] Kawasaki Heavy Industries

[g] Selective catalytic reduction of $NO_x$ is also applied

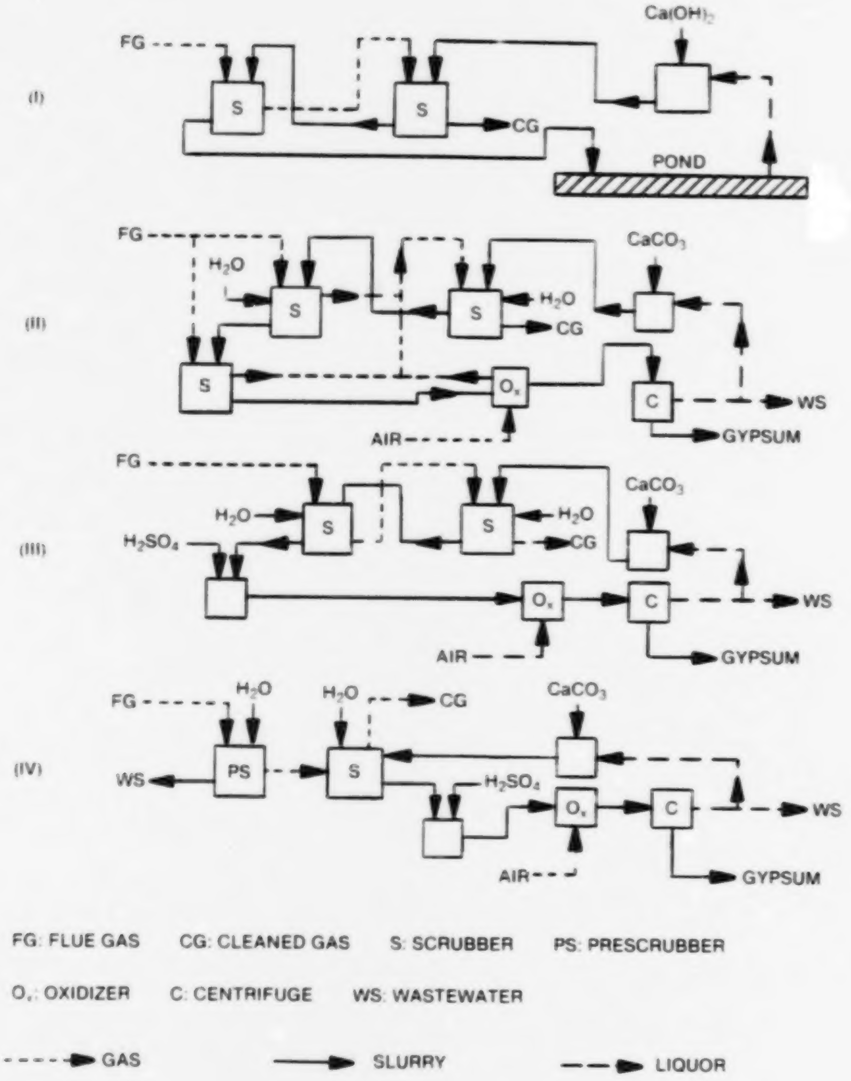

(I)
FG
S
S
CG
Ca(OH)$_2$
POND
(II)
FG
H$_2$O
S
S
H$_2$O
CG
CaCO$_3$
S
O$_x$
C
WS
AIR
GYPSUM
(III)
FG
H$_2$O
H$_2$SO$_4$
S
S
H$_2$O
CG
CaCO$_3$
O$_x$
C
WS
AIR
GYPSUM
(IV)
FG
H$_2$O
H$_2$O
CG
CaCO$_3$
WS
PS
S
H$_2$SO$_4$
O$_x$
C
WS
AIR
GYPSUM
FG: FLUE GAS
CG: CLEANED GAS
S: SCRUBBER
PS: PRESCRUBBER
O$_x$: OXIDIZER
C: CENTRIFUGE
WS: WASTEWATER
GAS
SLURRY
LIQUOR

**Figure 2-3** Types of Commercial Wet Lime/Limestone Processes for Coal.

For complete oxidation of calcium sulfite to gypsum, the pH of the sulfite slurry is usually lowered to about 4. Under the presence of a catalytic substance, complete oxidation is attained at a pH of about 5. Type II processes lower the pH to about 5 by using three scrubbers and a catalytic material for oxidation. The Type II system has been applied to 3 units, one for Mitsui Aluminum (No. 2 unit) and two for EPDC at Takasago.

Type III processes have two-stage scrubbers and lower the pH of the slurry from about 5 to about 4 by adding a small amount of sulfuric acid. This type requires a lower investment cost than does a Type II process, and may be preferred where sulfuric acid is available for a reasonable cost. Type III processes have been applied to four of EPDC's units, two units at Isogo constructed by Ishikawajima-Harima Heavy Industries (IHI) and two units at Takehara constructed by Babcock-Hitachi (BHK).

The Type IV process has a prescrubber and uses sulfuric acid for pH control. The prescrubber has the following functions. 1) It removes fly ash in the gas thus reducing the erosion problems of the scrubber system and increasing the purity of the gypsum. 2) It removes chloride and fluoride in the gas to protect the scrubber system from corrosion. The $SO_2$ removal efficiency as well as limestone utilization may be increased by the fluoride removal. It is likely that fluoride in combination with other impurities lowers the reactivity of limestone. 3) It cools the gas to help eliminate the scaling problem in the scrubber.

The Type IV process was developed and has been used by Mitsubishi Heavy Industries (MHI) since the 1960's. This technology has been applied to over 30 oil-fired boiler FGD units by MHI as well as to Chugoku Electric's coal-fired boilers at Shimonoseki in 1979 and Shin-Ube in 1982. Nearly all of the limestone scrubbing units constructed since 1979 use the Type IV process, including those being planned by IHI and BHK (Table 2-3).

In the U.S., prescrubbers with a separate liquor loop are seldom used because of the additional capital cost. The prescrubber treats a hot flue gas with an acidic liquor having a pH of about 1 and containing considerable amounts of chloride, fluoride, and fly ash. Therefore, the prescrubber must be constructed of highly corrosion- and erosion-resistant materials. For example, silicon carbide is often used for spray nozzles. On the other hand, the prescrubber substantially reduces the operational problems of the scrubber system. All of the new units using a prescrubber: the Shimonoseki unit (Section 4.2), the Tomato-Atsuma unit (Section 4.3), and the Matsushima units (Section 3.5), have been operated with 100% reliability since start-up (except for problems with the gas heating system at Shimonoseki). The two scrubber systems for the two 500 MW boilers at Matsushima are operated by only 2 persons per shift (one person/scrubber shift).

There are two limestone-gypsum units under construction that do not have a prescrubber with a separate liquor loop. One of them is EPDC's Takehara No. 3 unit which treats a flue gas that has gone through a highly efficient hot-side electrostatic precipitator with two spray towers in series and sulfuric acid for pH control. The other is Kyushu Electric's Minato unit which has a single spray tower for 90% $SO_2$ removal using a catalyst, and also sulfuric acid (Type III').

Shikoku Electric has two units under construction by Kawasaki Heavy Industries (Table 2-3). The units will use the magnesium gypsum process with a prescrubber with a separate liquor loop (Type IV'). For a coal-fired utility boiler Chiyoda may construct two FGD units which use the jet bubbling limestone-gypsum process and a prescrubber with a separate liquor loop.

Most U.S. systems do not have a prescrubber but have a stand-by scrubber which is not installed in Japan. The $SO_2$ removal efficiencies obtained with limestone scrubbing in the U.S. are not as high as those obtained with the Type IV system in Japan. The type IV system attains 95% removal without the use of system additives such as adipic acid which are widely used in the U.S.

A prescrubber may not be needed for throw-away sludge systems that can be operated in the unsaturated mode. But, for gypsum systems, the prescrubber is preferable particularly when high-quality gypsum is desired. Although the prescrubber increases the investment cost by 5 to 8 percent, the overall power generation cost, including that for FGD, may be lowered by such a highly reliable FGD system.

The Type IV process purges wastewater primarily from the prescrubber to remove fly ash, chloride, fluoride, etc., and also from the scrubber system to remove impurities derived from limestone. Types II and III purge wastewater from the scrubber system.

### 2.3.2 Scrubber Types and Operation Parameters

Table 2-4 shows the capacities, types of scrubbers, and operational parameters of FGD units for utility boilers presently in use in Japan. The EPDC's two units at Matsushima currently treat nearly 80% of the flue gas from the 500 MW boilers while Hokkaido Electric's unit at Tomato-Atsuma treats half of the flue gas from a 350 MW boiler. Other units treat all flue gases from the boilers. EPDC's Isogo and Takasago Stations have the same size boilers and almost identical FGD units.

EPDC's five FGD units at Isogo, Takasago, and Takehara were constructed between 1975 and 1977 and use venturi scrubbers or a combination of venturi and perforated plate scrubbers to attain a high dust removal efficiency. However, these units have a large pressure drop across the scrubbers (325-615 mm $H_2O$ in some cases) which results in high power consumption (2.2-2.6% of the power generated by the boiler). The relatively low power consumption at Isogo (2.2%) is due to its low inlet $SO_2$ concentration of 450 ppm.

The FGD units at Chugoku Electric's Shimonoseki Station were constructed in 1979 by MHI using the combination of a semi-venturi spray tower prescrubber and a plastic-grid packed tower. This configuration results in lower pressure drops and therefore relatively low power consumption (2.2%) than that associated with the EPDC units.

Hokkaido Electric's Tomato-Atsuma unit, constructed by Babcock Hitachi in 1980, uses an improved scrubber with a lower pressure drop and lower power

TABLE 2-4. OPERATION PARAMETERS OF FGD UNITS FOR COAL-FIRED UTILITY BOILERS (LIMESTONE-GYPSUM PROCESS)

| Station | EPDC Isogo | EPDC Takasago | EPDC Takehara | EPDC Matsushima | | Chugoku Shimonoseki | Hokkaido Tomato-Atsuma |
|---|---|---|---|---|---|---|---|
| Boiler No. | 1 | 1 | 1 | 1 | 2 | 1 | 1 |
| Capacity (MW) | 256 | 250 | 250 | 500x3/4 | 500x3/4 | 175 | 350x1/2 |
| Gas treated (1,000 Nm³/hr) | 821 | 792 | 793 | 1,300 | 1,300 | 610 | 610 |
| FGD constructor | IHI[a] | MM[b] | BHK[c] | IHI | BHK | MHI[d] | BHK |
| FGD start-up year | May '76 | Feb. '75 | Feb '77 | Jan. '81 | July '81 | July '79 | Oct. '80 |
| Inlet $SO_2$ (ppm) | 450 | 1,500 | 1,730 | 1,000 | 1,000 | 770 | 230 |
| Inlet dust (mg/Nm³) | 1,500 | 100 | 100 | 300 | 300 | 1000 | 25 |
| Ist scrubber (prescrubber) | Venturi | Venturi | Venturi | Spray | Spray | Spray | Venturi |
| L/G (liter/Nm³) | 7 | 6 | 2.5 | 3 | 3 | 3 | 2.2 |
| 2nd scrubber (scrubber) | Venturi | Venturi | PP[e] | Spray | Spray | Packed | PP[e] |
| L/G (liter/Nm³) | 7 | 6 | 7 | 15 | 15 | 14 | 5 |
| Outlet $SO^2$ (ppm) | 20 | 100 | 100 | 60 | 60 | 30 | 10 |
| Outlet dust (mg/Nm³) | 50 | 30 | 50 | 30 | 30 | 25 | 5 |
| $SO^2$ removal efficiency (%) | 95.5[f] | 93.3[f] | 94.2[f] | 94.0[g] | 94.0[g] | 96.1[g] | 95.7[g] |
| Dust removal efficiency (%) | 96.6 | 70.0 | 87.5 | 90.0 | 90.0 | 97.5 | 80.0 |
| Pressure drop (mm$H_2O$) | 360[h] | 325[h] | 615[h] | 145[i] | 133[i] | 240[h] | 450[j] |
| Wastewater (t/hr) | 10 | 7.5 | 12 | 15 | | 9 | 2.2 |
| Power requirement (%)[k] | 2.2 | 2.6 | 2.6 | 2.2 | | 2.2 | 1.7 |
| Gas reheating facility | OF[l] | OF[l] | OF[l] | GGH[m] | GGH[m] | GGH[m] | GGH[m] |

[a] Ishikawajima-Harima Heavy Industries
[b] Mitsui Miike Machinery Co.
[c] Babcock-Hitachi K. K.
[d] Mitsubishi Heavy Industries
[e] Perforated plate
[f] 1-3% lower after reheating by oil-firing
[g] 2-3% lower after gas reheating by gas-gas heater
[h] By two scrubbers and mist eliminators
[i] By two scrubbers
[j] Two scrubbers, mist eliminator, and gas-gas heater
[k] Percent of power generated
[l] Oil firing
[m] Gas-gas heater

consumption (1.7%) than that of the Takehara unit constructed by BHK in 1977 (2.6%). The low power consumption at Tomato-Atsuma is due to the improved FGD design as well as a low inlet $SO_2$ concentration of 230 ppm.

The two EPDC 500 MW boiler units at Matsushima were constructed in 1981 by IHI and BHK using spray towers with a low pressure drop (133-145 $mmH_2O$). The new unit for the EPDC Takehara No. 3 boiler under construction by IHI also uses a spray tower. The power consumption of the Matsushima No. 2 unit is 2.2%.

MHI used a spray tower absorber for the first lime-gypsum process unit (20 MW equivalent) constructed in 1964, but has used plastic grid-packed towers since 1972 after the scaling problem associated with these units had been solved. Plastic grid-packed towers generally provide greater removal efficiencies than spray towers. However, spray towers demonstrate a high efficiency when applied to scrubbers with stack heights of over 30 m, such as those recently constructed at Matsushima and Takehara (No. 3). The operation of those large units and three 600 MW units planned by Joban Joint Power and Hokkaido Electric (Table 2-3) which will use plastic grid-packed towers will provide a good comparison between the two tower types.

All of those units use limestone ground so that over 95% passes through a 325 mesh screen at a stoichiometry of 1.00 - 1.05 to remove 94-97% of inlet $SO_2$. The gas reheating adds a small amount of $SO_2$ so that the overall $SO_2$ removal efficiency is 90-95%. Oil-firing is used for reheating at Isogo, Takasago, and Takehara (No. 1); gas-firing (Ljungstrom heat exchanger) is used at the new units.

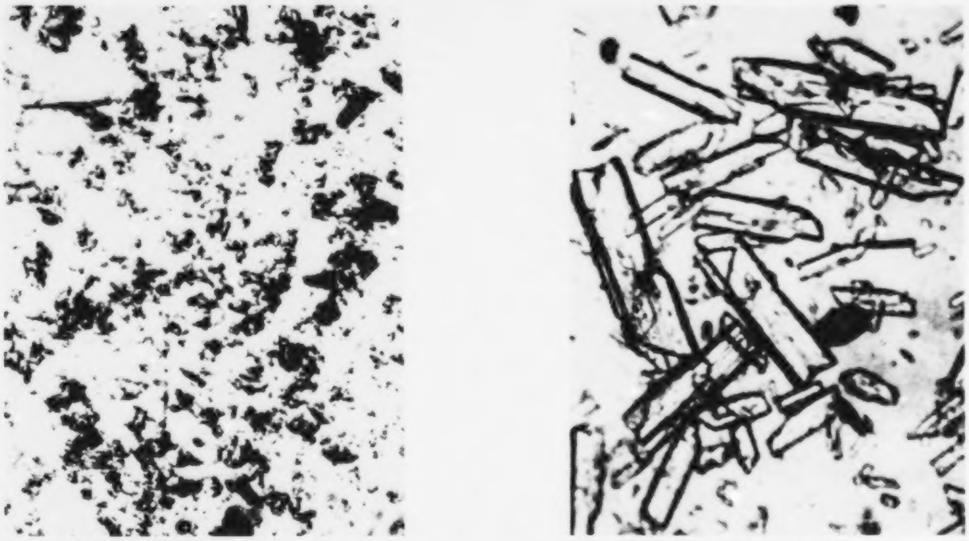
The dust removal efficiency of the scrubber systems ranges from 70 to 98%. The EPDC FGD units at Isogo and Chugoku Electric's unit at Shimonoseki have over 95% dust removal efficiencies because the boilers use low-sulfur coal for which the conventional cold-side ESP is not highly efficient. On the other hand, the EPDC boilers at Matsushima and Hokkaido Electric's boiler at Tomato-Atsuma all use hot-side ESPs which are efficient for low-sulfur coal, and in addition, the FGD units achieve dust removal efficiencies of 80-90%.

### 2.3.3 Scaling Prevention

The most significant operational problem associated with the lime/limestone FGD process has been the formation of gypsum scale in the scrubber and mist eliminator systems. In Japan these scaling problems have largely been solved, resulting in a reliability rate of more than 99% for lime/limestone FGD units.

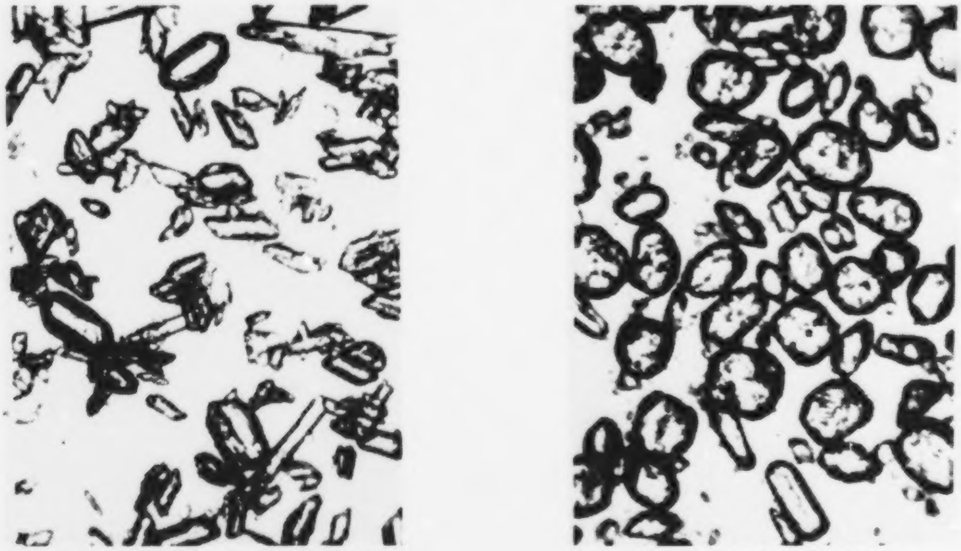
The No. 1 units at Omuta, Mitsui Aluminum, produce a throw-away sludge and are operated in the "unsaturated mode" keeping the oxidation ratio of calcium sulfite to sulfate below 10%. Since small amounts of sulfate can be contained in the crystal lattice of calcium sulfite, gypsum is not formed. Tests conducted by this author in 1975 and in 1981 confirm this phenomenon (Figure 2-4, G). The low oxidation ratio may be due to: 1) high inlet $SO_2$ concentrations (over 2,000 ppm) resulting in a relatively small $O_2/SO_2$ ratio, 2) a pH of over 7, and 3) possibly the oxidation inhibitor which may be present in carbide lime (7). When the sulfur content of the fuel is lowered, the oxidation ratio tends to increase, resulting in the formation of gypsum crystals in the scrubber system.

A number of methods originally developed in the 1960s by Mitsubishi Heavy Industries through basic studies and operation experience have been found to prevent scaling in gypsum producing processes. Seed crystals of gypsum can be added to the circulating slurry in the scrubber system to provide a site on which calcium and sulfate ions can deposit. This prevents the formation of gypsum crystals on the vessel surfaces (see Figure 2-4, H). Smooth structural material surfaces and continuous slurry flow in all parts of the scrubber system will prevent the deposit of gypsum crystals. Installation of a large slurry tank allows time in which the degree of gypsum supersaturation can be lowered. However, even with these countermeasures, gypsum scale tends to form on the mist eliminators. Washing the eliminators with fresh water or both fresh water and circulating liquor will dissolve gypsum deposits.



(A) PHOSPHOGYPSUM
DIHYDRATE PROCESS

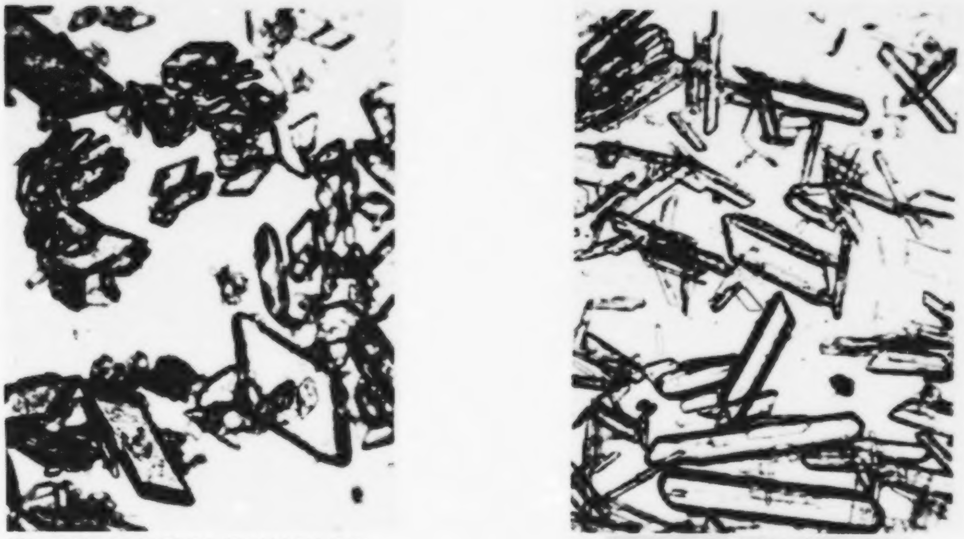
(B) PHOSPHOGYPSUM
HEMIHYDRATE-DIHYDRATE
PROCESS



(C) FGD GYPSUM, SHIMONOSEKI,
CHUGOKU ELECTRIC

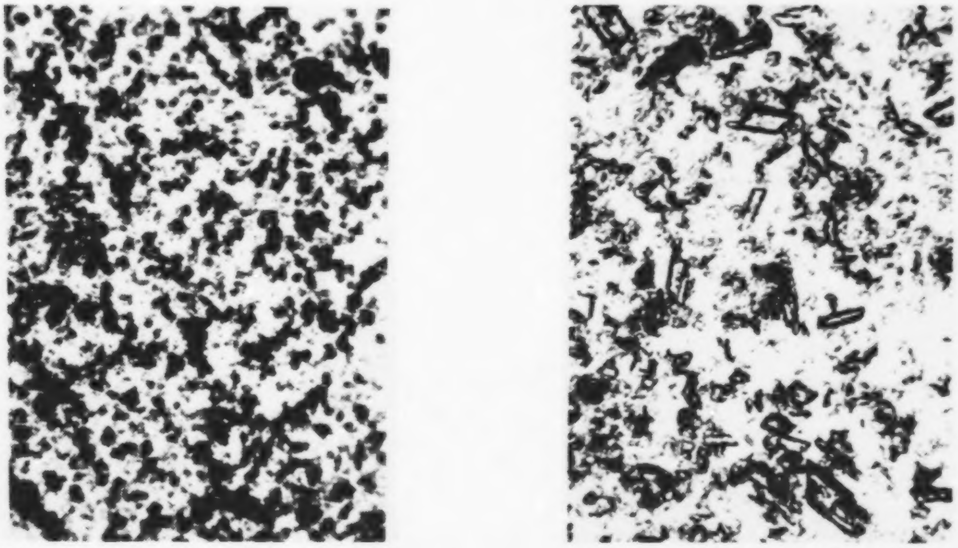
(D) FGD GYPSUM, TOMATO-ATSUMA, HOKAIDO
ELECTRIC

**Figure 2-4** Photomicrograms of By-Product Gypsum and Calcium Sulfite ( × 130)



(E) FGD GYPSUM, TAKASAGO, EPDC

(F) FGD GYPSUM, MgO-CaO PROCESS



(G) CALCIUM SULFITE SLUDGE OMUTA, MITSUI

(H) LIME SCRUBBING SCRUBBER OUTLET (CALCIUM SULFITE AND GYPSUM)

**Figure 2-4** (cont.) **Photomicrograms of By-Product Gypsum and Calcium Sulfite ( × 130)**

Next to scaling, corrosion is the most serious problem associated with FGD systems. This problem has been solved by using suitable materials and good lining fabrications. With these measures, FGD operations have become so reliable that not a single FGD unit in Japan has a stand-by scrubber, although most FGD units have stand-by pumps and centrifuges.

## 2.4 WASTEWATER AND ITS TREATMENT

### 2.4.1 Wastewater

A limestone-gypsum FGD unit for a 500 MW coal-fired boiler requires 40-80 tons/hr of water. Water is fed into the Type II and III process scrubbers and into both the prescrubber and scrubber of the Type IV. Some water also may enter the system as leakage from the pump-seals. Most of the water system is vaporized during the cooling of hot inlet gases, and 2-4 t/hr go into the gypsum byproduct through crystallization and moisture. To prevent the accumulation of impurities such as chloride and fluoride derived from coal, 5-30 tons/hr may be purged in the wastewater system. Flue gas from coal usually contains 30-300 ppm chloride and 10-100 ppm fluoride. Some other impurities are dithionate ($S_2O_6^{=}$) which forms in the scrubber, and magnesium, iron and manganese derived from fly ash and limestone.

### Type II and III systems

In the Type II and III FGD systems, chloride is caught by the scrubber and accumulates in the scrubber liquor. Since a high chloride concentration may cause stress corrosion, chloride concentrations in the liquor are usually kept at 5,000 ppm or below. The relationship between the chloride content of coal, wastewater purge rate, and the chloride content of the wastewater is shown in Figures 2-5 and 2-6.

As shown in Table 2-4, Type II and III FGD units (Isogo, Takasago, and Takehara of EPDC, 250-265 MW) purge 7.5 - 12 t/hr of wastewater. Actually, a portion of the supernatant of the gypsum slurry thickener is purged after being treated. This will be described in more detail later in this report.

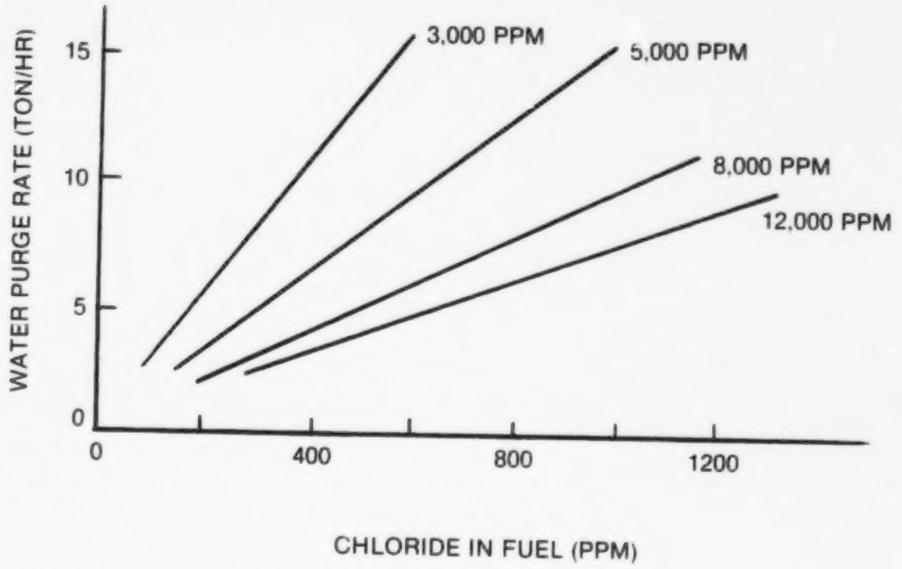


**Figure 2-5** **Relationship between Chloride Content of Fuel, Wastewater Purge Rate, and the Chloride Concentration in the Purge Stream.**

(250MW Boiler, 1,500ppm $SO_2$, 10% moisture in by-product gypsum.)

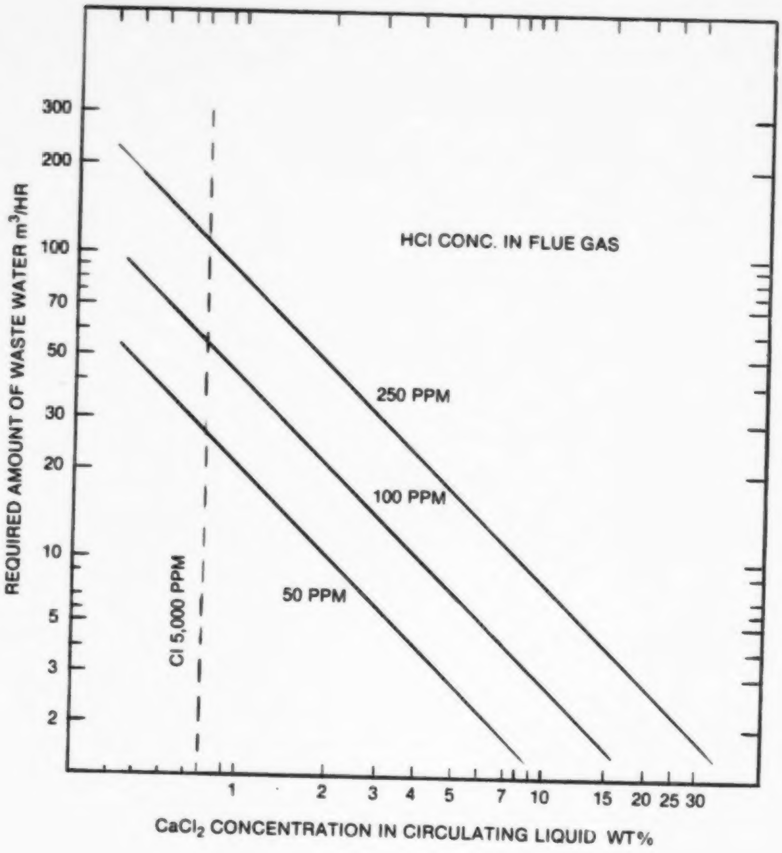


**Figure 2-6** Inlet HCl Concentration, $CaCl_2$ Concentration in Scrubber Liquor, and Amount of Wastewater for a 500MW Coal-fired Boiler. (Kobe Steel)*

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* For production of salable gypsum containing 10% moisture and 200ppm chloride.

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Most of the fluoride in the gas is caught by the scrubber liquor and forms calcium fluoride ($CaF_2$). The fluoride is in the form of very fine crystals and is not readily separated by precipitation or filtration. Therefore, a portion of it may be contained in the gypsum.

### Type IV system

With Type IV systems, most of the feed water is introduced into the prescrubber, where virtually all of the chloride and fluoride in the gas are caught. Most of the water is volatilized and the remainder is sent to wastewater treatment facilities. Main scrubber system water enters the wastewater system since fresh water must be used for the mist eliminator wash. The dithionate formed in the scrubber and impurities derived from the limestone are removed with the wastewater.

### 2.4.2 Wastewater Reduction

The wastewater from the Type IV process main scrubber system can be fed to the prescrubber only in a limited amount since it is saturated with gypsum; its evaporation in the prescrubber would form gypsum scale. At Hokkaido Electric's Tomato-Atsuma Station, most of the thickener supernatant is softened by adding sodium carbonate to precipitate calcium carbonate, and the softened water fed into the prescrubber. In this way, requirements for both fresh water and wastewater purging are considerably reduced; the wastewater produced is only 2.2 t/hr for a 175 MW unit (half of 350 MW). The new EPDC units at Matsushima and Takehara have similar systems. The Matsushima units, however, are not presently using the softening system and purge 15 tons/hr of wastewater per unit as mandated by government regulation.

Some regions in some countries, such as West Germany, do not allow the purging of wastewater containing certain concentrations of soluble salts, including calcium chloride. In such cases, it is important to minimize the amount of water which must be removed from the FGD system for evaporation and solids recovery. To achieve this, a scrubber system can be operated with a high chloride liquor using corrosion resistant materials. An FGD process such as the Kobe Steel process (Section 5.5) operating with 20-30% $CaCl_2$ liquor concentration may be useful for this purpose.

Figure 2-6 indicates that when the HCl content of flue gas is 100 ppm and the gas is treated with a 20% $CaCl_2$ solution to produce salable gypsum containing about 200 ppm Cl, the wastewater requirements are only 1.5 t/hr for a 500 MW coal-fired boiler. Wastewater requirements for flue gas containing 100 ppm HCl and 2,000 ppm $SO_2$ can be eliminated if gypsum containing about 1% Cl (10% liquor with 10% Cl) can be discarded.

Throw-away sludge systems using filtration and transportation of the cake remove a considerable amount of water from the system since a typical cake for this system contains about 60% liquid and 40% solids. The inlet and outlet chloride balances when the liquid in the cake contains 1-4% Cl.

### 2.4.3 Wastewater Treatment

The pH and composition of wastewater from commercial FGD units for coal-fired boilers in Japan are shown in Table 2-5. An example of a wastewater treatment system is shown in Figure 2-7. Wastewater from the prescrubber and scrubber is treated with lime to precipitate heavy metals and fluoride prior to filteration. The filtrate is then treated using an ion exchange resin procedure to remove dithionate $S_2O_6^{=}$ which can cause a chemical oxygen demand (COD). The small amount of concentrated dithionate solution obtained by desorption from the resin is either decomposed by adding an acid and oxidizing agent or by incineration.

Usually the treated water contains less than 10-15 mg/liter each of suspended solids and COD, less than 15 ppm fluoride, and less than 1 ppm oily material. The pH of the treated water ranges from 6.5 - 8.5 and thus satisfies wastewater treatment regulations.

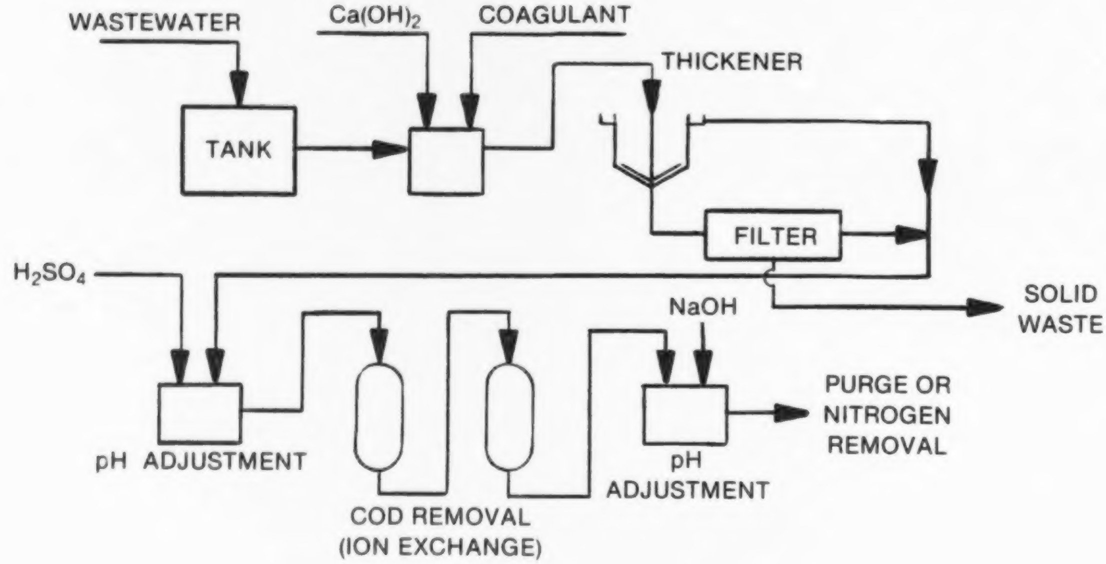

WASTEWATER
Ca(OH)2
COAGULANT
TANK
THICKENER
FILTER
H2SO4
NaOH
SOLID WASTE
PURGE OR NITROGEN REMOVAL
pH ADJUSTMENT
COD REMOVAL (ION EXCHANGE)
pH ADJUSTMENT

**Figure 2-7** **Typical FGD Wastewater Treatment System.**

In Japan there are no wastewater treatment regulations which control the content of soluble salts such as calcium and magnesium chlorides, since the treated water from the country's FGD units is discharged into the sea which is already rich in those soluble salts. If soluble salts must be controlled, the wastewater requirement should be eliminated in the manner described in Section 2.4.2.

In some power stations, a portion of the treated water is sprayed on the dry fly ash caught by ESPs. The water thereby controls fugitive dust emissions caused by handling operations or the wind.

As shown in Table 2-5, FGD wastewater contains a considerable amount of nitrogen compounds mainly in the form of nitrate derived from $NO_x$ in the flue gas. Flue gas from a coal-fired boiler usually contains 200 - 500 ppm $NO_x$. If 1-2% of the $NO_x$ is absorbed by the scrubbing liquor, the nitrogen content of the wastewater reaches 100 - 600 ppm for a 250 MW FGD unit purging 10 t/hr of wastewater (Table 2-5). To date, no removal treatment has been applied to nitrogen compounds, because their concentration is not considered to be a problem. Furthermore, abatement of $NO_x$ in the flue gas may reduce

TABLE 2-5. pH AND COMPOSITION (Mg/liter) OF WASTEWATER FROM FGD UNITS ON COAL-FIRED BOILERS PRIOR TO TREATMENT (ENVIRONMENT AGENCY)

| | | |
|---|---|---|
| pH | | 1 ~ 5 |
| Chemical oxygen demand | (COD) | 30 ~ 100 |
| Suspended solids | (SS) | 2,000 ~ 3,000 |
| Fluoride | (F) | 500 ~ 600 |
| Iron | (Fe) | 100 ~ 500 |
| Manganese | (Mn) | 10 ~ 30 |
| Magnesium | (Mg) | 200 ~ 1,000 |
| Aluminum | (Al) | 100 ~ 1,400 |
| Chloride | (Cl) | 3,000 ~ 8,000 |
| Total nitrogen[a] | (N) | 100 ~ 600 |

[a] When SRC $NO_x$ is not applied.

the nitrogen content in the wastewater. Application of SCR adds ammonia to the wastewater, but may not increase the total nitrogen content because it reduces nitrate (Section 2.7).

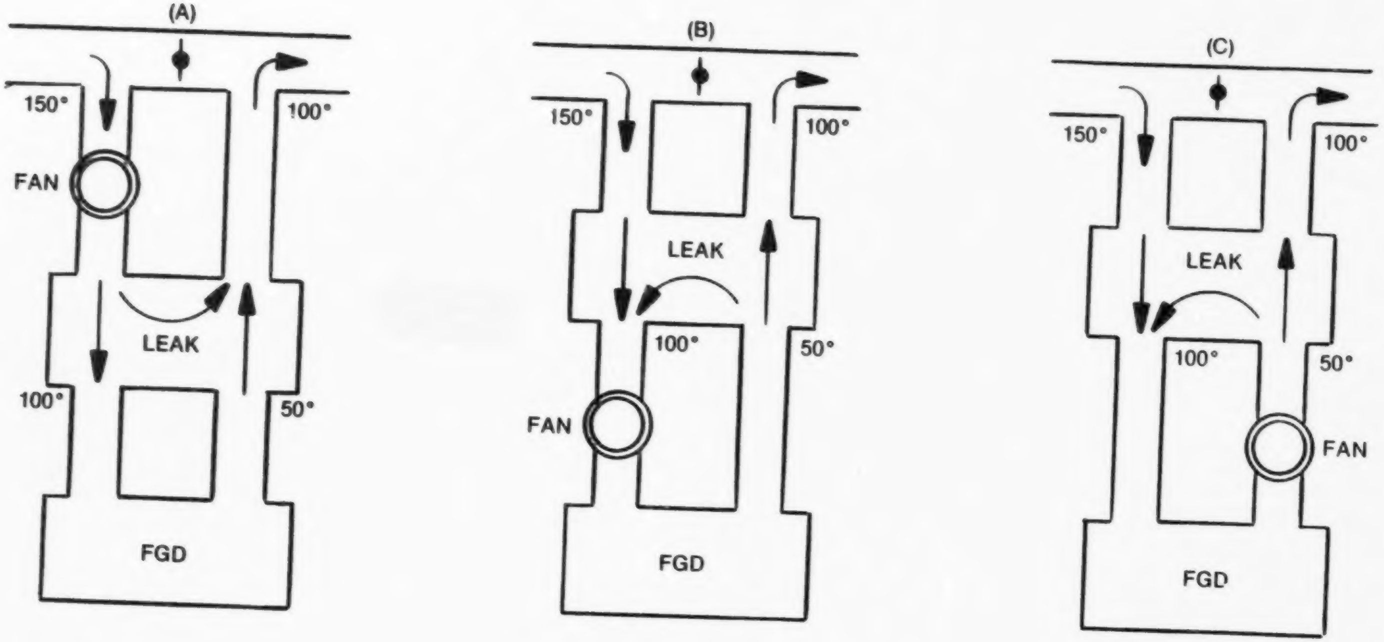
## 2.5 GAS-GAS HEATER (HEAT EXCHANGER)

In Japan, flue gas leaving wet process FGD systems at 50-60°C is usually heated to 90-110°C using low-sulfur oil. Since oil has become very costly, Ljungstrom (rotating) gas-gas heaters are now being used. This heater not only eliminates the fuel requirements but also cools the FGD inlet flue gas, thus reducing water consumption (Figure 2-8).

The major problem with the gas-gas heater is the deposition of solids within it. It is necessary to use a highly efficient mist eliminator to minimize these deposits. The first user of the gas-gas heater for a commercial FGD unit was Nippon Kokan at the Ogishima plant, Keihin Works. The heater was installed in an ammonia-scrubbing FGD system with a capacity of treating 1,120,000 $Nm^3$/hr of flue gas from an iron-ore sintering machine. At the Ogishima plant, the FGD outlet gas is first passed through a wet ESP in order to eliminate the ammonium sulfite-sulfate plume. Virtually all of the mists and dust in the flue gas are removed by the wet ESP, and the Ljungstrom unit has operated without problems.

However, some deposits may form in the gas-gas heater when a wet ESP is not used. Tests of gas-gas heaters with FGD systems for oil- and coal-fired boilers have indicated that the deposits associated with coal are much softer than those with oil, and can be removed by soot blowing. Even so, the commercial operation of the Ljungstrom gas-gas heater (for coal) has demonstrated that the heater is not void of dust deposits even with soot blowing. A 2 day water wash every 4-12 months is necessary to remove the dust. In addition, efforts have been made to reduce the amounts of deposition.

The first commercial gas-gas heater for a coal-fired boiler FGD system was installed in 1979 at Chugoku Electric's Shimonseki Station. All of the



(A)
150°
FAN
LEAK
100°
100°
50°
FGD
(B)
150°
100°
LEAK
100°
50°
FAN
FGD
(C)
150°
100°
LEAK
100°
50°
FAN
FGD

Figure 2-8 Rotating Gas-Gas Heater and Gas Leakage.

new FGD units for coal-fired boiler applications constructed after that time--Tomato-Atsuma, Matsushima, Shin-Ube, and Takehara (No. 3)--have used the gas-gas heater.

A problem associated with the rotating gas-gas heater operating at around 150°C is gas leakage. Usually a boost-up fan is placed upstream of the heater as shown in Figure 2-8 (A). In this case, the untreated gas leaks into the treated gas thereby lowering the removal efficiency. When the scrubber system's $SO_2$ removal efficiency is 95%, this gas leakage can lower that rate to about 90%.

Efforts have been made to reduce the leakage by modifying the design of the gas-gas heater. Placing the fan between the heater and FGD unit as shown in Figure 2-8 (B) and (C) causes leakage of treated gas into the untreated gas and results in little decrease in the removal efficiency. In this case, corrosion-resistant material is required for the fan because sulfuric acid may condense at the lower temperatures. The No. 2 FGD plant of Mitsui Aluminum (Section 4.1.3) uses system C, while EPDC's new unit at Takehara (No. 3) will use system B (8).

The cost of a rotating gas-gas heater is about 10% of the total capital cost of a limestone-gypsum FGD unit. However, the heaters account for a considerable fuel cost savings when gas heating of 50°C (from 50° to 100°C) is needed. When gas heating of 20-30°C is sufficient, simpler heating system such as the multi-tube type may be preferable.

## 2.6 BYPRODUCT GYPSUM

### 2.6.1 Demand, Supply, and Price

Gypsum has been the major byproduct of FGD in Japan. Since Japan has little natural gypsum, there has been a market for virtually all of this byproduct material. Gypsum supply and demand are shown in Figure 2-9. Yearly gypsum demand had reached about 6 million tons by 1980. The major demand is for cement setting retarder and wallboard; the major source is phosphogypsum,

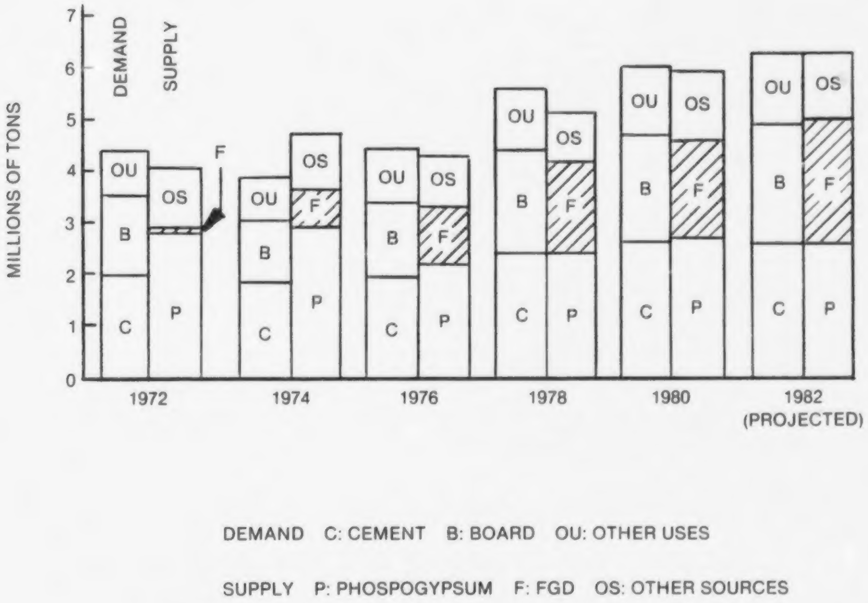


Figure 2-9 Gypsum Supply and Demand in Japan.

a byproduct of wet process phosphoric acid production with phosphate rock and sulfuric acid. In 1972, the gypsum supply was limited and the price of byproduct gypsum reached 3,000 yen/ton (Figure 2-2). In 1974, gypsum was in over supply because of the rapid increase in FGD gypsum systems and the decreased demand due to an economic recession. These factors resulted in a decrease in the price of byproduct gypsum to almost zero. The price has increased since 1976, and again has reached an average of 3,000 yen/ton (Figure 2-2).

Although the return from gypsum sales offsets only a small portion (around 5%) of the annualized FGD cost, it is a good idea to sell gypsum in Japan because of the limited land space available for storage or disposal.

### 2.6.2 Crystal Size and Quality

Phosphogypsum obtained by the conventional dihydrate process popular in the U.S. consists of small crystals about 10 μm in size (Figure 2-4, A). Phosphogypsum contains about 20% moisture after filtration and a considerable amount of impurities such as phosphoric acid and fluorine. These impurities decrease the quality of gypsum to the point that it must usually discarded as waste.

The hemihydrate-dihydrate process for phosphoric acid production has been developed in Japan in order to produce a salable quality of gypsum. This gypsum consists of large crystals, 30-200 μm in size (Figure 2-4, B), and contains fewer impurities than that produced by the conventional process. It contains about 10% moisture after filtration, and has been used for cement, wallboard, plaster, and other products.

FGD byproduct gypsum has been accepted by users in Japan without difficulty because it contains no harmful impurities such as phosphoric acid. Gypsum-producing FGD units in Japan are designed to obtain a good quality gypsum. Examples of FGD gypsum are shown in Figure 2-4, C through F; the size of most crystals ranges from 30 to 200 μm.

After being centrifuged, gypsum from limestone scrubbing usually contains 7-10% moisture and looks almost dry. A gypsum moisture content of greater than 10% is not desirable for two reasons. First, when used as a cement setting retarder, moisture-rich gypsum tends to form bridges in the hoppers and cannot be charged smoothly to the cement mills. Second, for wall-board or plaster applications, additional energy must be expended for drying moist gypsum.

The size and thickness of the crystals is important in determining gypsum quality. Gypsum often grows in thin plate or needle-like crystals which not only hold moisture but also diminish the strength needed for wallboard and plaster. Gypsum crystals produced by Type IV limestone scrubbing processes tend to grow into grain-like shapes (Figure 2-4, C and D) which are not very large but are fairly thick. The grainy gypsum has a small surface area and low moisture content (7-8%) giving it good strength characteristics.

Magnesium derived from lime or limestone or added to improve lime scrubbing tends to depress the growth of gypsum crystals. Nevertheless, good quality gypsum can be obtained by using a suitable design and properly operating the FGD unit (Figure 2-4, F).

### 2.6.3 Effects of Impurities on Gypsum Quality

The major impurities found in FGD-produced gypsum are:

(1) Unreacted lime or limestone

(2) Calcium sulfite

(3) Fly ash and unburned carbon

(4) Chloride and fluoride

(5) Ammonium and sodium

## Unreacted lime and limestone

The amount of unreacted lime or limestone in gypsum is related to the $CaO/SO_2$ or $CaCO_3/SO_2$ mole ratio and the $SO_2$ removal efficiency. In Japan, a mole ratio of 0.95-0.05 is normally used to remove 90-97% of the $SO_2$. This means that the weight ratio of lime or limestone to gypsum may be 2-4 to 100. For example, the gypsum obtained at EPDC's Takasago unit contains 2-3% limestone. The gypsum produced by FGD systems Types III or IV contains virtually no limestone, because sulfuric acid is added to the scrubber outlet slurry in order to lower the pH.

Unreacted lime present in FGD gypsum raises the pH of the gypsum and makes it unsuitable for wallboard production. When the pH of gypsum exceeds 10, setting is delayed, strength is lowered, and its adhesibility to paper is reduced (9). Gypsum which contains unreacted lime may not be suitable for cement because free lime may cause the cement to set improperly. On the other hand, the effects of unreactive limestone are less severe. A fairly large amount of limestone in gypsum produces no noticeable effect on cement, although it may affect wallboard or plaster characteristics. Most users of FGD gypsum have set no specifications on limestone content since it is usually very low. One user of gypsum for cement specifies that the limestone content should be below 1%. The specification, however, may not be necessary.

## Calcium sulfite

The calcium sulfite content of FGD gypsum in Japan is below 1% because the sulfite is well oxidized. Calcium sulfite consits of very small crystals (below 5 μm as shown in Figure 2-4, G), and tends to hold large amounts of moisture. Since calcium sulfite is not reactive, a 10% sulfite concentration in gypsum produces no adverse effect on cement and wallboard properties except for the problems associated with increased moisture content.

### Fly ash and unburned carbon

The fly ash content in byproduct gypsum is usually below 5% and at this level has no appreciable effects on wallboard properties. For cement applications, fly ash content produces no problems, even in large amounts, except for increasing the moisture content of the product. The gypsum at EPDC's Isogo Station contains about 20% fly ash and 15% moisture and has been used in the marufacture of a special building material.

The unburned carbon content in fly ash is usually below 5% and at this level has no adverse effect on cement. The carbon occasionally exceeds 5% due to poor control of combustion modification measures designed to reduce $NO_x$. The carbon absorbs the AE (air entraining) agent sometimes used in very small amounts to improve the quality of concrete. A large amount of unburned carbon is not desirable when an AE agent is used.

### Chloride and fluoride

The scrubber liquor of Type II and III FGD systems contains about 5,000 ppm of chloride. Since byproduct gypsum usually contains about 8% moisture, the chloride content of the gypsum is generally about 400 ppm. This amount of chloride is usually acceptable for various uses of gypsum. If further reduction of chloride is needed, gypsum can be washed with fresh water in the centrifuge. With type IV systems, the chloride content of the scrubber liquor is usually below 1,000 ppm and the content of the gypsum byproduct is below 100 ppm.

The fluoride content is much lower than the chloride content in byproduct gypsum. Moreover, calcium fluoride is insoluble. Therefore, fluoride generally does not affect the quality of gypsum. In some cases the scrubber liquor contains a considerable amount of fluoride which acts to lower the reactivity of the limestone and thus increases the limestone content of the gypsum.

### Magnesium, ammonium and sodium

Magnesium is derived from lime or limestone and tends to accumulate in the scrubber liquor; in some processes, magnesium is added to lime to improve the scrubbing. When present, magnesium tends to hinder the crystal growth of gypsum. However, with proper design of the reactor or crystallizer, a good quality gypsum can be obtained even with the presence of a large concentration of magnesium (Figure 2-4, F, Section 5.2).

When SCR is applied upstream, the scrubber liquor may contain ammonia. The ammonia content of the scrubber liquor, however, does not usually exceed 500 ppm, and at this level does not affect gypsum quality. In some cases, a small amount of ammonium sulfate is added to gypsum as a setting promotor.

The sodium content of a wet limestone process scrubber liquor is usually very low. On the other hand, gypsum from sodium-lime or sodium-limestone double alkali processes contains an appreciable amount of sodium when the gypsum is not washed with fresh water. It has been shown that when gypsum contains a 0.3% concentration of sodium sulfate, its setting tiem is shortened and its strength is weakened.

In general, the impurities in byproduct gypsum produced from wet limestone processes are found in small amounts and do not affect the gypsum's quality. When byproduct gypsum is in oversupply, its users tend to set strict specifications on the impurity content, but when gypsum demand is high, they readily user lower-grade material. The presence of a considerable amount of impurities may change the properties of gypsum appreciably. However, most impure gypsum is still useful as long as the amount of impurities remains farily constant.

## 2.7 SELECTIVE CATALYTIC REDUCTION COMBINED WITH FGD

### 2.7.1 Selective Catalytic Reduction (SCR)

Ammonia selectively reacts with $NO_x$ in gases at 150-450°C in the presence of a catalyst and oxygen. SCR can be conveniently applied to boiler economizer outlet gas at 300-400°C:

$$4NO + 4NH_3 + O_2 = 4N_2 + 6H_2O$$

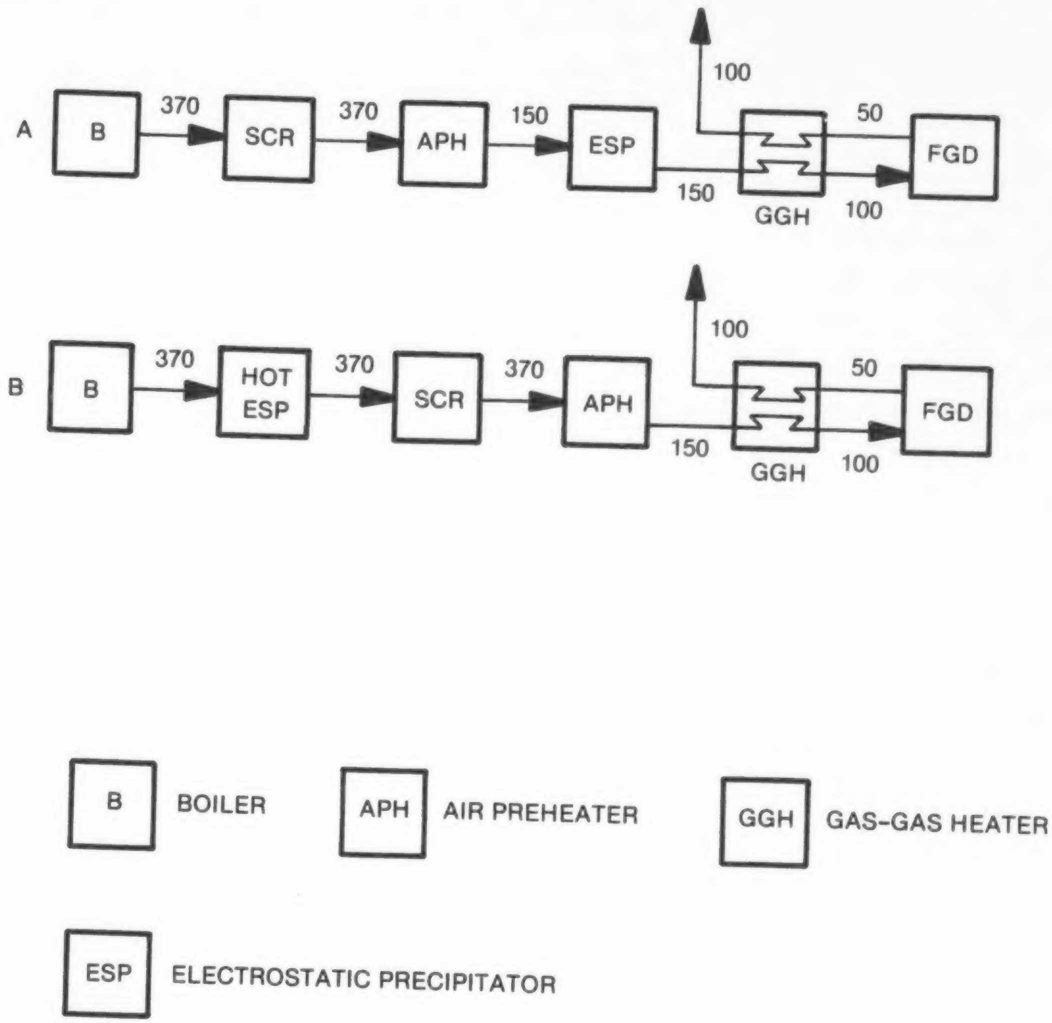
Although SCR had some serious problems during the initial stages of its development, these have been solved. SCR is now a simple operation, and an SCR unit is usually operated automatically (without an operator).

### 2.7.2 Combined SCR/FGD Systems

Figure 2-10 shows two combined SCR-FGD systems for coal-fired boilers. The SCR reactor is system A treats flue gas leaving a cold ESP with a full dust load of about 20 g/Nm³ (high-dust system). The reactor in system B treats the gas leaving a hot EPS which contains less than 0.1 g/Nm³ of dust (low-dust system). The high-dust system has been used at the Shimonoseki and Shin-Ube plants of Chugoku Electric, while the low-dust system is used at EPDC's Takehara plant and the Tomato-Atsuma plant of Hokkaido Electric. Both systems have been operated successfully without erosion or plugging of the catalyst by fly ash.

With the high-dust system, a small amount of ammonia--0.01-0.02% for 3-7 ppm of unreacted ammonia--may be contained in the fly ash caught by the cold ESP. However, in general, fly ash containing less than 0.02% ammonia presents no problems when used for cement additive or for land fill.

Ammonia leaving the SCR reactor is caught by a wet FGD system. Because ammonia at the scrubber outlet is virtually nonexistent, the usual plume problem experienced at ammonia scrubbing FGD units does not exist.


A
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SCR
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SCR
370
APH
100
150
GGH
100
50
FGD
B
BOILER
APH
AIR PREHEATER
GGH
GAS-GAS HEATER
ESP
ELECTROSTATIC PRECIPITATOR

**Figure 2-10** **SCR-FGD Combination Systems (Figures show gas temperatures, °C).**

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### 2.7.3 Ammonia in Scrubber Liquor and Wastewater

Ammonia at a concentration of 5 ppm in flue gas from a 250 MW coal-fired boiler amounts to about 3 kg/hr. If the FGD unit for the boiler purges 10 t/hr of wastewater, the scrubber (or prescrubber) liquor as well as the wastewater may contain about 300 mg/l of ammonia. The ammonia does not affect the scrubber operation but may influence wastewater treatment.

Adding lime to wastewater for heavy metal precipitation causes much of the ammonia to be volatilized. If necessary, even more ammonia can be removed from the water. EPDC's demonstration plant at Takehara (combined SCR and Type II FGD) has a unit to remove ammonia from wastewater using a conventional activated sludge process; ammonia is reduced to less than 10 mg/liter.

It is likely that the nitrogen compounds shown in Table 2-5 will be reduced considerably by applying SCR and reducing $NO_x$ in flue gas. It is possible that the total nitrogen compounds (including ammonia) may be lowered by applying SCR and keeping unreacted ammonia at a very low level; e.g., 1 ppm or below at the Shimonoseki plant.

## 2.8 ECONOMICS OF THE LIMESTONE-GYPSUM FGD PROCESS

### 2.8.1 Investment Costs

The investment costs of FGD units using limestone-gypsum processes for coal-fired boilers are listed in Table 2-6. The cost of the first units completed in 1975-1976 was 18,600 yen/kW. The investment cost has since continued to increase due mainly to inflation. Chuguku Electric's Shimonoseki plant is fairly expensive because it includes a gas-gas heater to reheat the gas to 110-120°C. The gas-gas heater accounts for about 10% of the investment cost but eliminates fuel oil expenses needed for gas heating and thus provides an overall economic advantage.

TABLE 2-6. INVESTMENT COST OF FGD UNITS FOR COAL-FIRED BOILERS (LIMESTONE-GYPSUM PROCESS, RETROFIT)

| Power Company | Plant Site | Capacity (MW) | Investment Cost | | Year of Completion |
|---|---|---|---|---|---|
| | | | $10^9$ yen | yen/kWhr | |
| EPDC | Takasago | 250 + 250 | 9.3 | 18,600 | 1975-1976 |
| EPDC | Isogo | 265 + 265 | 11.0 | 20,750 | 1976 |
| EPDC | Takehara | 250 | 7.2 | 28,800 | 1977 |
| Chugoku Electric | Shimonoseki | 175[a] | 6.0 | 34,300 | 1979 |

[a] Including Ljungstrom type gas-gas heat exchanger

The estimated FGD cost for a 500 MW coal-fired boiler equipped with a gas-gas heater is shown in Table 2-7. The annualized FGD cost is about 1.9 yen/kWhr. About 70% of the cost is made up of the fixed cost with a 7 year depreciation schedule. When the annualized cost is based on a 15-20 year depreciation schedule, as is commonly done in the U.S., the result is a considerably lower yen figure.

### 2.8.2 Power Generation Cost

A rough estimated cost for power generation is shown in Table 2-8. About 20% of the total investment for a coal-fired power station is allocated for flue gas treatment consisting of FGD, SCR and ESP. The investment cost of FGD accounts for about 70% of the cost for flue gas treatment. The estimated

TABLE 2-7. ESTIMATED FGD COST IN 1981 (Assuming a new 500 MW coal-fired boiler, limestone-gypsum process, 70% boiler utilization, 3,066,000 MWhr/year, Inlet $SO_2$ 1,000 ppm, 90% $SO_2$ removal, 7 years depreciation, 10% interest.)

| | |
|---|---|
| Investment Cost (including gas-gas heater) | 16 billion yen<br>(32,000 yen/kW) |
| Annual Cost (millions of yen) | |
| Fixed Cost (25% of investment) | 4,000 |
| Power (2.1% of power generated, 17 yen/kWhr) | 1,100 |
| Labor (13 persons) | 100 |
| Limestone (43,000 t) and chemicals | 250 |
| Others (maintenance, etc.) | 500 |
| Total | 5,950 |
| Gypsum (84,000 t, ¥2500/t) | - 210 |
| Total | 5,740 |
| Annualized Cost | 1.87 yen/kWh |

annualized cost of power generation for a coal-fired power station is 18 yen/kWhr, of which nearly three yen/kWhr goes for flue gas treatment. The estimated annualized power cost using low-sulfur oil (64,000 yen/kl containing 0.27% sulfur) with an electrostatic precipitator is about 21 yen/kWhr including 15 yen/kWhr for the fuel cost.

TABLE 2-8. ESTIMATED POWER GENERATION COST FOR A NEW 1000 MW COAL-FIRED BOILER (1981)

| | |
|---|---|
| Coal-fired Boiler | |
| Investment cost (yen/kW) | |
| Power station excluding pollution control facilities | 210,000 |
| Flue gas desulfurization (90% efficiency) | 30,000 |
| Selective catalytic reduction (80% efficiency) | 7,000 |
| Electrostatic precipitator (99.5% efficiency) | 4,000 |
| Other pollution control facilities including ash disposal | 17,000 |
| Total | 268,000 |
| Annualized cost (yen/kWhr) | |
| Flue gas desulfurization | 1.7 |
| Selective catalystic reduction | 0.7 |
| Electrostatic precipitator | 0.2 |
| Other pollution control including ash disposal | 1.7 |
| Fuel (Coal at 15,000 yen/ton) | 6.4 |
| Others | 7.3 |
| Total | 18.0 |
| Oil-fired Boiler | |
| Annualized power cost with low-sulfur oil at 64,000 yen/kl with electrostatic precipitator (yen/kWhr)* | 21.0 |

*Including 7 years depreciation for pollution control facilities and 15 years depreciation for power generation facilities.

# SECTION 3

# FGD UNITS OF THE ELECTRIC POWER DEVELOPMENT CO. (EPDC)

## 3.1 OVERVIEW

The Electric Power Development Company (EPDC) was established in 1952 by the Japanese Government jointly with nine major power companies for the purpose of developing electric power. Consumption of domestic coal became one of the major objectives of EPDC in the 1960s when most power companies changed fuel from coal to oil. EPDC currently is the largest coal consumer for power generation.

EPDC's boilers and FGD units are listed in Tables 1-6 and 2-2, respectively. Before EPDC installed the first FGD units in 1975, it conducted an extensive survey of various FGD processes. Since over 90% $SO_2$ removal was not easily attainable using the dry process, the survey focused on wet processes. EPDC's evaluation of wet processes is shown in Table 3-1. Because any FGD process with a "bad" or "worse" rating is not desirable for use with utility boilers, EPDC selected the wet limestone-gypsum process. Scrubbers developed in the U.S., such as Chemico's venturi and Babcock & Wilcox's venturi/perforated plate combination, were chosen because the Japanese FGD process developers had little experience with flue gas from coal-fired boilers at that time. Japanese licensees (Mitsui Miiki Machinery Co. and Ishikawajima-Harima Heavy Industries for Chemico and Babcock Hitachi Ltd. for Babcock & Wilcox) constructed the FGD units.

As shown in Tables 3-2 and 3-3, all of the units constructed at Isogo, Takasago, and Takehara have operated with very high reliability: 100% for Isogo, over 98% for Takasago, and over 99% for Takehara. When an FGD unit is down, the boiler usually switches fuel to low-sulfur oil in order to

TABLE 3-1. PRE-EVALUATION OF WET FGD PROCESSES(8)

| Evaluation Item | | Process | | | | | Cost | |
|---|---|---|---|---|---|---|---|---|
| Name of Process | | Desulfurization Efficiency | Simplicity | Secondary Pollution | Ease of Operation | Actual Use | Construction | Operation |
| Throw-Away Process | (1) Calcium Base | o | ● | Δ | ● | o | ● | o |
| | (2) Sodium Base | ● | ● | Δ | ● | o | ● | Δ |
| Gypsum Recovery Process | (3) Lime-Limestone | o | o | o | o | ● | o | o |
| | (4) Double Alkali | ● | Δ | o | Δ | o | Δ | o |
| | (5) Dilute Sulfuric Acid | o | o | o | o | o | Δ | Δ |
| | (6) Ammonia-Lime | ● | Δ | Δ | Δ | Δ | o | Δ |
| Sulfuric Acid Recovery Process | (7) Sodium Base | ● | x | o | x | o | x | o |
| | (8) Magnesium Base | ● | x | ● | x | o | x | o |

| Remarks: | | |
|---|---|---|
| | ● | Best |
| | o | Good |
| | Δ | Bad |
| | x | Worse |

TABLE 3-2. ACTUAL HOURS OF EPDC'S PLANT AND FGD OPERATIONS: 1974-1977 (8)
(Unit: Hour)

| Plant | Unit | 1974 |  | 1975 |  | 1976 |  | 1977 |  | Total |  | Operability |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
|  |  | A | B | A | B | A | B | A | B | A | B |  |
| Isogo | 1 |  |  | 676 | 676 | 7699 | 7699 | 8115 | 8115 | 16490 | 16490 | 100% |
|  | 2 |  |  |  |  | 7031 | 7031 | 7641 | 7641 | 14672 | 14672 | 100% |
|  | Total |  |  | 676 | 676 | 14730 | 14730 | 15756 | 15756 | 31162 | 31162 | 100% |
| Takasago | 1 | 975 | 957 | 8206 | 8053 | 7515 | 7438 | 8381 | 8328 | 25077 | 24776 | 98.7% |
|  | 2 |  |  |  |  | 8143 | 8008 | 7703 | 7595 | 15846 | 15603 | 98.4% |
|  | Total | 975 | 957 | 8206 | 8053 | 15523 | 15446 | 16084 | 15923 | 40923 | 40399 | 98.6% |
| Takehara | 1 |  |  |  |  | 1056 | 1056 | 7895 | 7895 | 8951 | 8951 | 100% |
| Total | - | 975 | 957 | 8924 | 8729 | 31741 | 31252 | 39735 | 39574 | 81036 | 80512 | 99.3% |

Year 1974 = April 1, 1974 - March 31, 1975
A = hours of boiler operations
B = hours of FGD operation

TABLE 3-3. OPERATION HOURS OF EPDC'S BOILERS AND FGD PLANTS: 1978-1981

| | | 1978 | | 1979 | | 1980 | | 1981 | | Total | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | A | B | A | B | A | B | A | B | A | B | B/A(%) |
| Isogo | No. 1 | 7704 | 7704 | 8045 | 8043 | 7694 | 7694 | 8188 | 8188 | 31906 | 31904 | 100.0 |
| | No. 2 | 8206 | 8206 | 7820 | 7820 | 7935 | 7935 | 7691 | 7691 | 31652 | 31652 | 100.0 |
| Takasago | No. 1 | 7829 | 7823 | 8041 | 8035 | 7736 | 7736 | 8282 | 8270 | 31888 | 31864 | 99.9 |
| | No. 2 | 8166 | 8147 | 8013 | 8006 | 8048 | 8048 | 7234 | 7200 | 31461 | 31401 | 99.8 |
| Takehara | No. 1 | 7583 | 7583 | 8161 | 8161 | 8134 | 8134 | 7304 | 7304 | 31182 | 31182 | 100.0 |
| Matsushima | No. 1 | | | | | 1784 | 1784 | 7204 | 7204 | 8988 | 8988 | 100.0 |
| | No. 2 | | | | | | | 6754 | 6754 | 6754 | 6754 | 100.0 |

A Boiler operation hours

B FGD operation hours

continue operating without FGD. Therefore, the B/A ratio in Tables 3-2 and 3-3 also shows the reliability of the FGD unit. All of the units remove over 90% of $SO_2$.

There are four major problems associated with these FGD units. These are: 1) high pressure drop resulting in considerable power consumption (Table 2-3); 2) problems caused by impurities in the flue gas such as fly ash, chloride and fluoride; 3) consumption of a large amount of water and purge of wastewater; and 4) the energy requirement for gas reheating.

To solve the first problem, a low pressure drop scrubber (spray tower) is used at the new units at Matsushima and Takehara (No. 3). To eliminate the problem of impurities in the flue gas, the Matsushima units use a Type IV system (Figure 2-3) instead of Types II and III. To minimize the water problems, the new units all have a gas-gas heater (heat exchanger) and also a softening facility for the blowdown water from the scrubber system (Section 3.5). To solve problems (3) and (4) entirely, EPDC has conducted tests of a dry FGD system which uses activated carbon and will construct a demonstration plant for further testing in 1984 (Section 3.6).

## 3.2 FGD UNITS AT TAKASAGO

### 3.2.1 Process Description

EPDC's first FGD unit (Figure 3-1) was constructed at the Takasago Station by Mitsui Miiki Machinery Co. and went into operation in 1975 to treat flue gas from the existing No. 1 boiler (250 MW). A second similar unit began operation on the No. 2 boiler (250 MW) in 1976. Specifications and major equipment are shown in Tables 3-4 and 3-5, respectively.

The Takasago boilers use coal containing about 2% sulfur. At the ESP exit, the 150°C flue gas contains about 1,500 ppm $SO_2$ and 100 mg/$Nm^3$ of dust. The scrubber was designed to reduce the $SO_2$ concentration in flue gas

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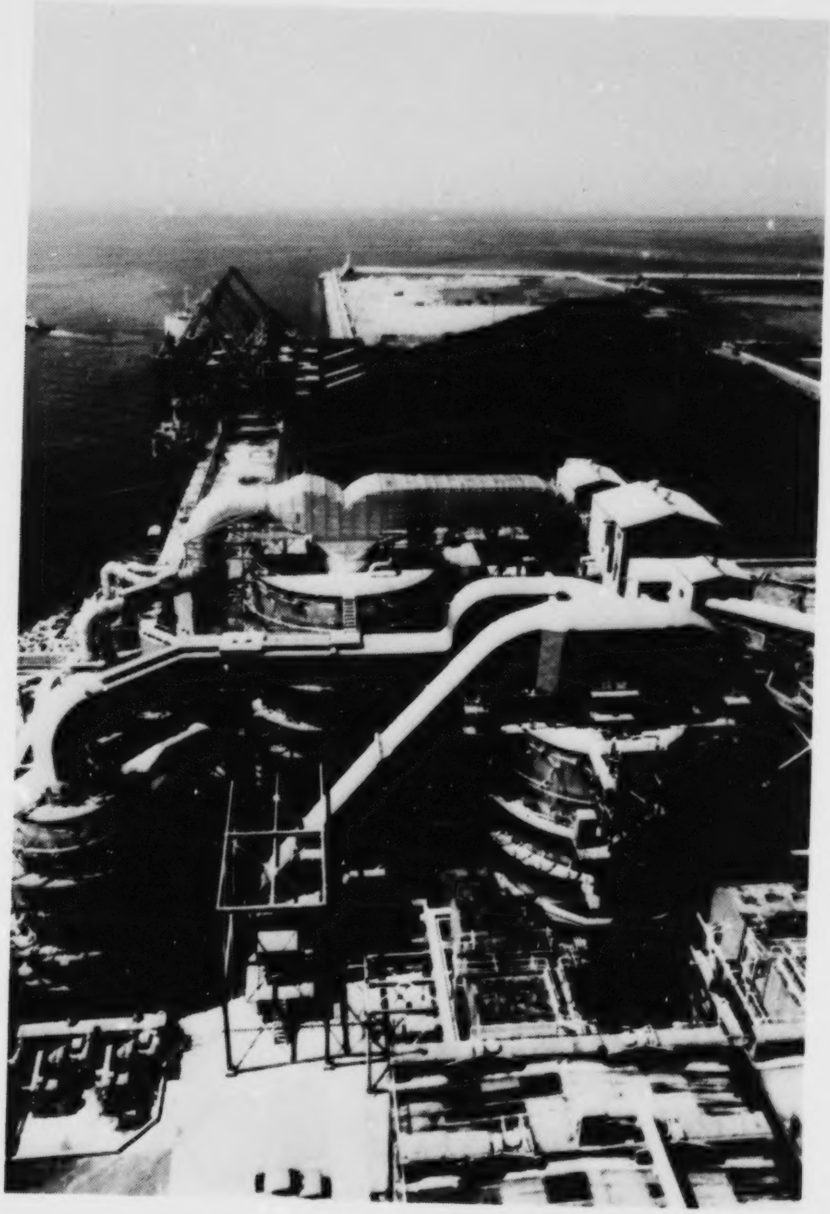

Figure 3-1 FGD Unit at Takasago, EPDC (No. 1)

## TABLE 3-4. SPECIFICATIONS OF FGD UNITS (TAKASAGO)

| | Designed | Actual |
|---|---|---|
| Power generating capacity, MW | 250 | |
| Fuel consumption, tons/hr | 89.4 | |
| Inlet gas volume, dry, $10^3$ $Nm^3/hr$ | 743 | 799 |
| $O_2$ in gas, % | 4.7 | 5.5 - 6.0 |
| Gas temperature, °C | 140 | 150 - 160 |
| $SO_2$ inlet concentration, ppm | 1500 | 1400 - 1600 |
| $SO_2$ (scrubber outlet), ppm | below 100* | 70 - 80 |
| $SO_2$ removal efficiency, % | 93.3 | 95** |
| Inlet dust, $mg/Nm^3$ | 100 | 80 |
| Outlet dust, $mg/Nm^3$ | below 50* | 20 |
| Requirements | | |
| Water, ton/hr | 46 | 52 |
| Limestone, ton/hr | 5.1 | |
| Power, kW | 8000 | 6500 |
| Wastewater, ton/hr | | 7.5 |

* Guaranteed by Mitsui Miike

** About 91% after gas reheating.

## TABLE 3-5. MAJOR EQUIPMENT OF TAKASAGO NO. 1 UNIT

| Equipment | Type and Dimensions |
|---|---|
| Limestone tanks | 1000 t x 2 |
| Absorber (1st) | Venturi 11.5 m ϕ x 20 m |
| Absorber (2nd) | Venturi (double throat) 14 m ϕ x 23 m |
| Forced draft fans | (12,300 $m^3/min$, 1900 kW) x 2 |
| Circulation pumps | |
| For 1st absorber | 135 kW x 4 |
| For 2nd absorber | 200 kW x 5 |
| pH adjusting tower | Venturi 6 m ϕ x 16 m |
| Reactors (oxidizers) | 6 m ϕ x 18 m, 2 units |
| Blowers for oxidizers | (110 $Nm^3/hr$, 1000 mm $H_2O$, 250 kW) x 3 |
| Centrifuges | (solid 1.5 t/hr) x 7 |
| Reheating furnaces | (oil 625 liter/hr) x 4 |

below 100 ppm to give an overall $SO_2$ removal efficiency of 90%. This would occur after the flue gas is reheated, thereby increasing the $SO_2$ concentration to about 140 ppm.

A flowsheet of the FGD process used at Takasago is shown in Figure 3-2. The unit includes two venturi scrubbers (Chemico) and another venturi for pH control of the slurry. For oxidation of calcium sulfite to gypsum, the No. 1 unit has two reactors as shown in Figure 3-2 and Table 3-5, while the No. 2 unit has just one.

Limestone with over 98% purity ground to pass through a 325 mesh screen is used in the scrubber. A water-soluble (inorganic compound) catalyst is added to the limestone slurry to promote $SO_2$ absorption and oxidation of calcium sulfite to gypsum. The limestone slurry with a stoichiometry of almost 1.0 is fed into the second scrubber. The slurry leaving the second absorber is fed into the first absorber, while a major portion of flue gas is introduced into the first scrubber and then into the second one. In the second scrubber, the pH of the slurry is 6.0-6.3 and the L/G ratio 6-7 liter/$Nm^3$, while in the first scrubber, the pH is 5.6-6.1 and the L/G 5.5-6.0.

After leaving the first scrubber, the slurry is fed into the pH control tower, where it is contacted with a small portion of the flue gas in order to lower the pH to 5.4-5.8. The slurry then goes into the reactor (oxidizer), where it is again contacted with a small portion of flue gas and air for further pH reduction and oxidation of calcium sulfite to gypsum. All of the gas leaving the pH controller and the reactor is sent to the second absorber in order to attain over 90% $SO_2$ removal. The 55°C treated gas is heated to 80-85°C by oil-firing and sent to a stack.

The gypsum slurry leaving the reactor in 5-7% concentration is treated in a thickener until it reaches a 20% concentration and then centrifuged to yield a salable gypsum with 6-8% moisture. A portion of filtrate from the centrifuge is purged from the system at a rate of 7.5 tons/hr to maintain a 5,000 ppm or lower chloride concentration in the scrubber liquor. The

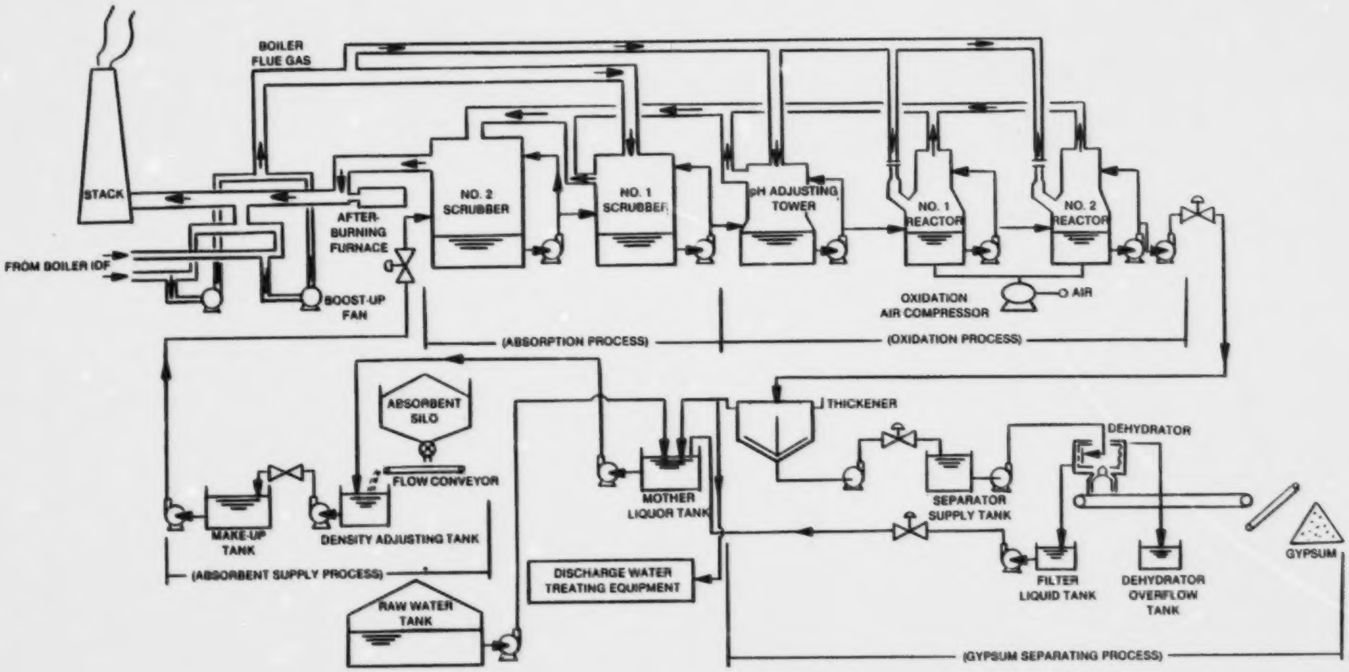
BOILER
FLUE GAS
STACK
FROM BOILER IDF
AFTER-
BURNING
FURNACE
BOOST-UP
FAN
NO. 2
SCRUBBER
NO. 1
SCRUBBER
pH ADJUSTING
TOWER
NO. 1
REACTOR
NO. 2
REACTOR
OXIDATION
AIR COMPRESSOR
AIR
(ABSORPTION PROCESS)
(OXIDATION PROCESS)
ABSORBENT
SILO
FLOW CONVEYOR
MAKE-UP
TANK
DENSITY ADJUSTING TANK
(ABSORBENT SUPPLY PROCESS)
RAW WATER
TANK
MOTHER
LIQUOR TANK
DISCHARGE WATER
TREATING EQUIPMENT
THICKENER
SEPARATOR
SUPPLY TANK
DEHYDRATOR
FILTER
LIQUID TANK
DEHYDRATOR
OVERFLOW
TANK
GYPSUM
(GYPSUM SEPARATING PROCESS)

Figure 3-2 Flowsheet of FGD Unit No. 1, Takasago.

purged water is treated as described in Section 2.4.3. The catalyst is purged with wastewater, which is neither hazardous nor costly. In order to prevent corrosion, a plastic lining is used in most of the vessels and pipings.

### 3.2.2 Performance

Initially, the chevron mist eliminators in the scrubbers were washed with circulation liquor. Since this liquor is saturated with gypsum, scaling occurred on the eliminators, which necessitated periodic shutdowns of the FGD units for cleaning. The average reliability of the units from the beginning of operation until 1977 was below 99% (Table 3-2) primarily due to this problem. As a result, the washing system was altered so that the eliminators now are washed with both fresh water and circulation liquor. The scaling problem thus has been solved and the units have had over 99% reliability since 1978 as shown in Tables 3-3, 3-6, and 3-7.

The boilers are operated at a reduced load at night. A schematic of the load changes and the gas compositions is shown in Figure 3-3. At full load, the $SO_2$ concentration at the scrubber outlet is kept slightly below 100 ppm (about 94% removal). Since the reheating process uses oil, the $SO_2$ concentration increases to about 130 ppm, producing an overall removal efficiency of about 91%. Normally, the boiler load is changed at a rate of 3 MW/min. Higher $SO_2$ removal efficiency is attained at a low load.

The daily requirements for operating one FGD unit at Takasago are as follows.

$CaCO_3$ - 118 tons

Power - 136,000 kWh

Oil - 41 liters (reheating)

Steam - 13 tons (heating of oil)

TABLE 3-6. PERFORMANCE OF FGD UNIT AT TAKASAGO (NO. 1)

| Month | Hours<br>Total (A) | Hours<br>Boiler operation (B) | Hours<br>FGD operation (C) | Hours<br>FGD called to operate (D) | Utilization (%)<br>Boiler (B/A) | Utilization (%)<br>FGD (C/A) | Operability (C/B, %) | Reliability (C/D, %) | SO₂ removal (%) |
|---|---|---|---|---|---|---|---|---|---|
| April 1980 | | | | | | | | | |
| May | 2184 | 1688[a] | 1688 | 1688 | 77.3 | 77.3 | 100 | 100 | 92 |
| June | | | | | | | | | |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.0 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.8 |
| September | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 93.0 |
| October | 744 | 744 | 739[b] | 744 | 100 | 99.3 | 99.3 | 99.3 | 92.7 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 92.7 |
| December | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.7 |
| January | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.5 |
| February | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 92.6 |
| March | 744 | 216[a] | 216 | 216 | 29.0 | 29.0 | 100 | 100 | 92.6 |
| April | 720 | 242[a] | 242 | 242 | 33.6 | 33.6 | 100 | 100 | 92.8 |
| May | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.1 |
| June | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 92.8 |

---

[a] Boiler was shut down for annual maintenance.

[b] FGD was shut down for 4 hours for maintenance.

TABLE 3-7. PERFORMANCE OF FGD UNIT AT TAKASAGO (No. 2)

| Month | Hours | | | | Utilization (%) | | Oper-ability (C/B, %) | Reli-ability (C/D, %) | $SO_2$ removal (%) |
|---|---|---|---|---|---|---|---|---|---|
| | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | | | |
| April 1980 | | | | | | | | | |
| May | 2184 | 1641[a] | 1641 | 1641 | 75.1 | 75.1 | 100 | 100 | 92 |
| June | | | | | | | | | |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.8 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.8 |
| September | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 93.1 |
| October | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.6 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 92.7 |
| December | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.6 |
| January 1981 | 744 | 670 | 670 | 670 | 90.0 | 90.0 | 100 | 100 | 92.4 |
| February | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 92.5 |
| March | 744 | 744 | 733 | 733 | 100 | 100 | 98.5 | 100 | 92.4 |
| April | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 92.8 |
| May | 744 | 264[a] | 264 | 264 | 35.9 | 35.9 | 100 | 100 | 93.0 |
| June | 720 | 29[a] | 29 | 29 | 4.0 | 4.0 | 100 | 100 | |

[a] Boiler was shut down for annual maintenance.

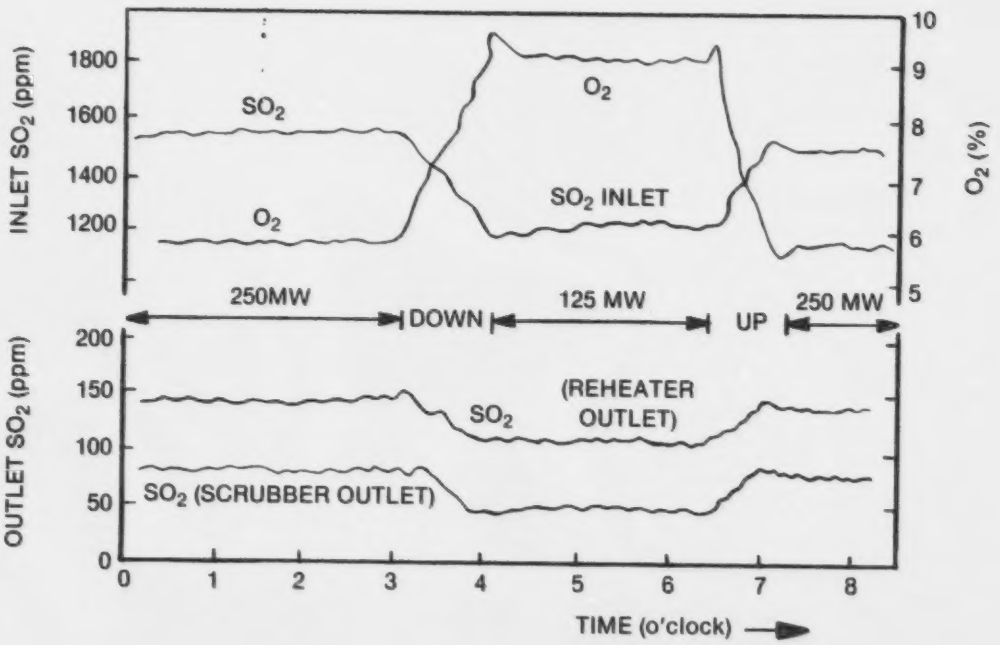

**Figure 3-3** Change in Gas Concentrations with Boiler Load, Takasago.

70A2494

Industrial water - 1,200 tons

Catalyst - 80,000 yen

The power requirement accounts for about 2.6% of the power generated by the boiler. The investment cost for the two FGD units was 9.3 billion yen in 1976. Three people per shift operate the units with another three people working in the daytime for maintenance.

### 3.2.3 Evaluation

The FGD units all have successfully removed over 90% of $SO_2$ with nearly 100% reliability. The venturi scrubber is efficient for dust removal but not highly efficient for $SO_2$ removal and causes a considerable pressure drop and power consumption. For a boiler with a highly efficient dust removal facility, low pressure drop scrubbers, such as spray tower and grid-packed tower, may be preferable. With those scrubbers, over 90% $SO_2$ removal can be attained using limestone without a catalyst.

In order to promote oxidation, the Mitsui Miike process uses flue gas to lower the pH of the slurry. This system does not require sulfuric acid but does require additional scrubbers. In locations where sulfuric acid is available for a reasonable cost, using acid is simpler and requires a lower investment cost and therefore may be preferable.

## 3.3 FGD UNITS AT ISOGO

### 3.3.1 Overview

The Isogo power station is located in Yokohama City adjacent to Tokyo and has two 265 MW coal-fired boilers. Because of its location, the most stringent air quality regulations apply to the plant. For $SO_2$ abatement, the station originally used domestic low-sulfur coal containing 0.3 - 0.4% sulfur and ESP. Since further reduction of $SO_2$ and particulates was required, FGD units using the limestone-gypsum process were installed on the two boilers in 1976 (Figure 3-4). The units were constructed by Ishikawajima-Harima Heavy Industries. Since electrostatic precipitators are not very efficient for use with low-sulfur coal, Chemico venturi scrubbers are used to remove over 90% of particulates and $SO_2$.

The specifications of the boiler, ESP, and FGD unit are shown in Table 3-8. Since the installation of the FGD units, the boiler has used a blend of domestic coals with 0.6% sulfur. The flue gas leaving the ESP contains about 1,500 mg/$Nm^3$ of dust with about 450 ppm $SO_2$. Over 90% of the dust and $SO_2$ are removed by the FGD system.

The $NO_x$ concentration in the flue gas has been lowered to 150-190 ppm by combustion modification using a low-$NO_x$ burner and staged combustion. Since FGD units were installed in a small space beside the boiler, there is no additional room to install a reactor for selective catalytic reduction (SCR) of $NO_x$.

### 3.3.2 Process Description

The primary equipment and a flowsheet of the FGD unit used at Isogo are shown in Table 3-9 and Figure 3-5. Limestone ground finely enough to pass through a 325 mesh is used as the absorbent. The limestone slurry is fed into the No. 2 scrubber, where the pH of the circulating slurry is maintained at 6.0. The slurry is then sent to the No. 1 scrubber where the pH is kept at 4.5. Flue gas is first fed into the No. 1 scrubber, where 40-60% of the $SO_2$ is absorbed, and then to the No. 2 scrubber, where another 55-35% of the $SO_2$ is absorbed. This results in an overall $SO_2$ removal efficiency of 95%.

The slurry discharged from the No. 1 scrubber is sent to a pH adjusting tank, where sulfuric acid is added to lower the pH to about 4.0. The slurry is then fed into an oxidizer, where calcium sulfite is oxidized to gypsum using air bubbles.

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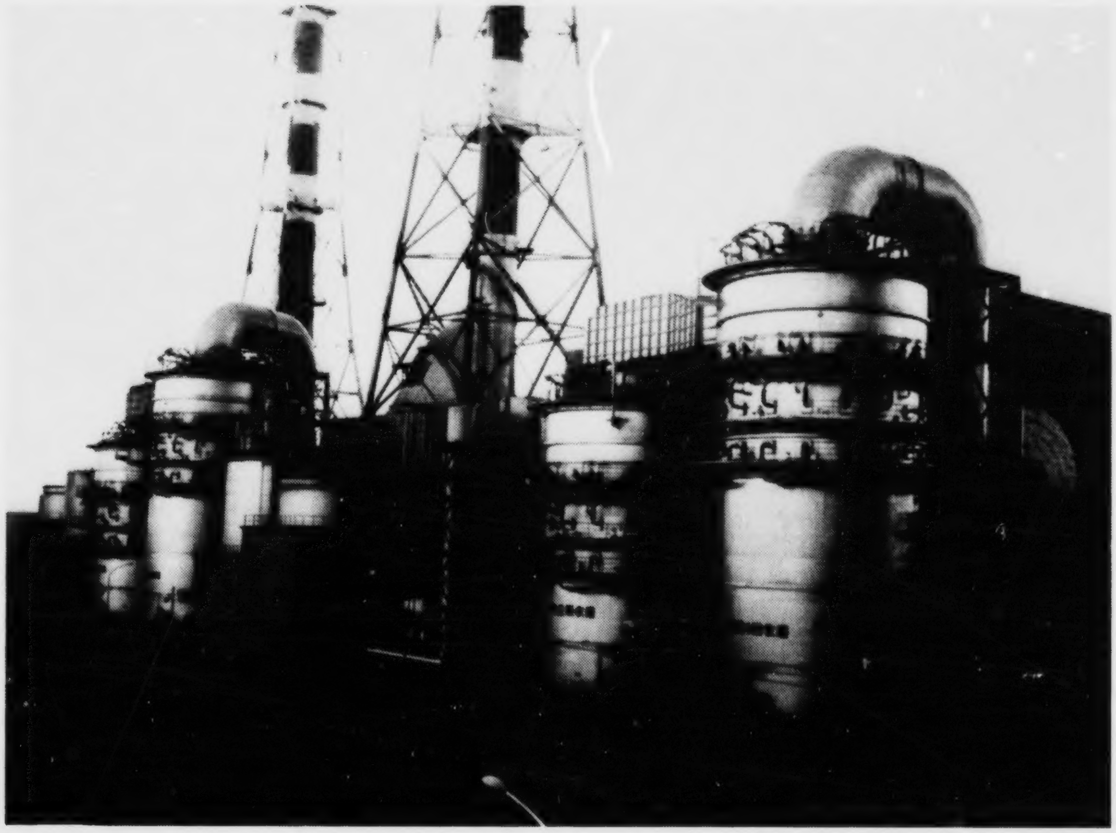

Figure 3-4 Isogo Plant, EPDC (No. 1 Unit Left, No. 2 Unit Right)

TABLE 3-8. PLANT SPECIFICATIONS (ISOGO)

| | |
|---|---|
| **Boiler (265 MW)** | |
| Type | FW Single Drum Radiation Reheat, Natural Circulation and Indoor |
| Evaporation Rate, T/hr | 840 |
| Flue Gas Volume, $Nm^3/hr$ | 879,100 (MCR) |
| Coal Composition | Inherent Water 4.2%<br>Ash Content 16.0%<br>Calorific Value 6,200 kcal/kg<br>Sulfur Content 0.3 – 0.6% |
| Flue Gas Temperature, °C | 143 (airheater outlet) |
| Draft Control | Balanced Draft |
| Manufacturer | Ishikawajima-Harima Heavy Industries Co., Ltd. |
| **Dust Collector** | |
| Type | Cold-side ESP |
| Efficiency, % | 98 |
| **Flue Gas Desulfurization System** | |
| Type | IHI-CHEMICO Type, Limestone Slurry Scrubbing/Gypsum Recovery |
| Gas Volume, $Nm^3/hr$ | 900,000 (MCR)<br>450,000 (night hours) |
| Efficiency of $SO_2$ Removal, % | 90 |
| Efficiency of Dust Removal, % | 96.7 |
| Absorbent, T/D | Limestone: 50 |
| By-Product, T/D | Gypsum: Approximately 120 |
| Water Requirement, T/hr | 80 |
| Wastewater, T/hr | 10 |
| Gas Reheating System | Oil-Firing |
| Total Area of Installation, $m^2$ | Approximately 8,000 |
| Manufacturer | Ishikawajima-Harima Heavy Industries Co., Ltd. |

TABLE 3-9. EQUIPMENT (ISOGO)

| | |
|---|---|
| **Absorbent Supply Process** | |
| Limestone Silo, t | 1,000 (common use for No. 1 and No. 2 Boilers) |
| Pump for $CaCO_3$ Slurry | 3 Sets |
| **Absorption Process** | |
| 1st Stage Absorber | 1 Stage Venturi Scrubber 1 Set<br>SS Flake Lining<br>13.3 m (dia.) x 30.9 m (height) |
| 2nd Stage Absorber | 1 Stage Venturi Scrubber 1 Set<br>SS Flake Lining<br>13.3 m (dia.) x 23.4 m (height) |
| Boost Up Fan | Dual Suction Turbo-Fan Direct Coupled To Motor Shaft |
| Gas Volume, $m^3$/min | 13,400 each |
| Draft, mm $H_2O$ | 820 |
| Motor Power, kW | 2,400 x 2 Sets/Unit |
| Circulation Pump, kW | 210 x 5/Unit 195 x 5/Unit |
| **Gypsum Production Process** | |
| Oxidation Tower Feed Tank | Vertical Cylindrical Type Made of Carbon Steel - 1 Set |
| Oxidation Tower | Vertical Cylindrical Type Made of Carbon Steel - 2 Sets/Unit |
| Oxidation Blower | Roots Blower |
| Air Volume, $Nm^3$/min | 50 each |
| Draft, mm $H_2O$ | 20,000 |
| Motor Power, kW | 200 x 3 Sets/2 Units |
| Thickener | Sedimentation and Concentration Type - 1 Set (6 m in dia.) |
| Dehydrator | Screw Decanter Type |
| Solid Content, T/hr | 1.05 x 4 Sets/Unit |
| Gas Reheater | Horizontal Cylindrical Type |
| After-Burning Furnace, kg/hr | 2,600 - 1 Set |

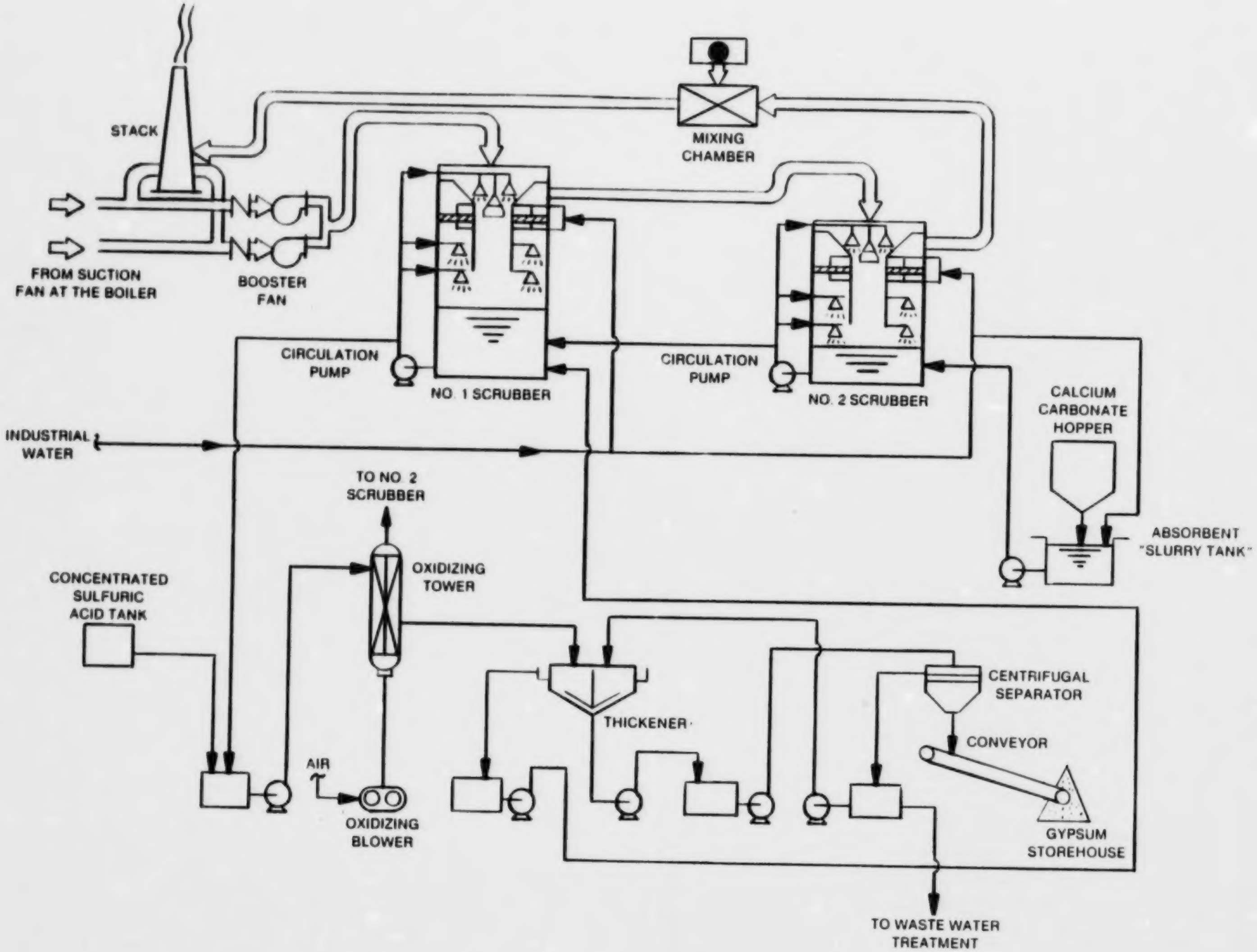
STACK
MIXING CHAMBER
FROM SUCTION FAN AT THE BOILER
BOOSTER FAN
CIRCULATION PUMP
NO. 1 SCRUBBER
CIRCULATION PUMP
NO. 2 SCRUBBER
CALCIUM CARBONATE HOPPER
INDUSTRIAL WATER
TO NO. 2 SCRUBBER
ABSORBENT "SLURRY TANK"
OXIDIZING TOWER
CONCENTRATED SULFURIC ACID TANK
CENTRIFUGAL SEPARATOR
THICKENER
CONVEYOR
AIR
OXIDIZING BLOWER
GYPSUM STOREHOUSE
TO WASTE WATER TREATMENT

Figure 3-5 Flowsheet of FGD Unit, Isogo.

The gypsum slurry is sent to superdecanters, to a continuous centrifuge and through a thickener. The byproduct gypsum has a 15% moisture content and contains about 15% fly ash. Although it is fairly impure and has a high moisture content, the gypsum can still be used to produce a building material, as long as its composition does not fluctuate too much.

Most of the liquor discharged from the superdecanters is sent to the No. 1 scrubber, while a portion (10 t/hr/unit) is purged to prevent the accumulation of chloride in the circulating liquor; the chloride concentration is kept at a concentration of 5,000 ppm. The wastewater undergoes standard treatment as described in Section 2.4.3.

### 3.3.3 Performance and Economics

The FGD units at Isogo have been operated with 100% reliability since startup, as shown in Tables 3-2, 3-3, 3-10, and 3-11. Although there have been minor troubles with superdecanters and slurry recirculation pumps, maintenance generally is performed while the FGD plants are in operation. Batch centrifuge may be preferred to the superdecanter to prevent problems. The FGD plant with two units has been operated by 3 persons per shift. In the daytime, one more person is assigned to each unit for chemical analysis and maintenance.

The $SO_2$ removal efficiency of the units ranges from 95% to 97% at the scrubber outlet. When oil firing is used to reheat the gas, a small amount of $SO_2$ is added, but nonetheless the overall $SO_2$ removal efficiency still exceeds 90%. When the units originally began operation, limestone slurry was prepared by using a circulating liquor. Since it has been shown that the use of the circulating slurry lowers the reactivity of limestone, fresh water now is used for the slurry preparation. In order to retain the high $SO_2$ removal efficiency, a small amount of chemical has been added to the slurry.

The Isogo plant with its two FGD units cost 11 billion yen in 1976. The boilers have been operated at full load in the daytime and at about half

TABLE 3-10. PERFORMANCE OF FGD UNIT AT ISOGO (No. 1)

| Month | Hours |  |  |  | Utilization (%) |  | Oper-ability (C/B, %) | Reli-ability (C/D, %) | $SO_2$ removal (%) |
|---|---|---|---|---|---|---|---|---|---|
|  | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) |  |  |  |
| April 1980<br>May<br>June | 2184 | 1185[a] | 1185 | 1185 | 54.2 | 54.2 | 100 | 100 | 94 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.3 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.7 |
| September | 720 | 718[b] | 718 | 718 | 99.7 | 99.7 | 100 | 100 | 96.2 |
| October | 744 | 680 | 680 | 680 | 91.4 | 91.4 | 100 | 100 | 96.0 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 95.5 |
| December | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.0 |
| January 1981 | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 97.2 |
| February | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 96.1 |
| March | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.2 |
| April | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 95.1 |
| May | 744 | 720[a] | 720 | 720 | 97.3 | 97.3 | 100 | 100 | 96.7 |
| June | 720 | 176[a] | 176 | 176 | 24.4 | 24.4 | 100 | 100 |  |

[a] Boiler was shut down for annual maintenance.

[b] Boiler was shut down for 2 hours to change the voltage of the transformer.

## TABLE 3-11. PERFORMANCE OF FGD UNIT AT ISOGO (No. 2)

| Month | Hours |  |  |  | Utilization (%) |  | Operability (C/B, %) | Reliability (C/D, %) | $SO_2$ removal (%) |
|---|---|---|---|---|---|---|---|---|---|
|  | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) |  |  |  |
| April 1980<br>May<br>June | 2184 | 1689[a] | 1689 | 1689 | 77.3 | 77.3 | 100 | 100 | 94 |
| July | 744 | 656[b] | 656 | 656 | 88.1 | 88.1 | 100 | 100 | 94.9 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 96.0 |
| September | 720 | 718 | 718 | 718 | 99.7 | 99.7 | 100 | 100 | 95.7 |
| October | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.3 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 95.2 |
| December | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.0 |
| January 1981 | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 95.7 |
| February | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 97.1 |
| March | 744 | 504 | 504 | 504 | 67.7 | 67.7 | 100 | 100 | 95.5 |
| April | 720 | 0 | 0 | 0 |  |  |  |  |  |
| May | 744 | 534 | 534 | 534 | 71.8 | 71.8 | 100 | 100 | 97.5 |
| June | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 95.8 |

[a] Boiler was shut down for annual maintenance.

[b] Boiler was shut down for repair.

load at night. The two units daily consume about 100 tons of limestone and produce about 240 tons of gypsum. In addition, the plant consumes about 7 tons/day of 35% NaOH liquor and 0.7 ton/day of 98% $H_2SO_4$. The power consumption of the FGD units is about 2.2 percent of the total power generated by the boiler.

### 3.3.4 Evaluation

The 100 percent reliability rate for five years of operation is an excellent record and may be the result of good design, fabrication and operation. The low inlet $SO_2$ concentration and extensive monitoring by the local government, using a telemeter-computer system, also may have contributed to this excellent record.

The reduced reactivity of limestone when the circulation liquor is used may be due to the effect of the fluoride in the liquor. Fluoride reacts with limestone to form a coating of insoluble calcium fluoride. The coating may not form in the scrubber because limestone's reaction with $SO_2$ produces $CO_2$ which prevents the formation of coating. The reduction in limestone reactivity has not been significant in the FGD units at Takasago and Takehara. However, at Isogo, an especially high limestone reactivity is needed because of the requirement that 95% of the $SO_2$ be removed using the venturi scrubber which is not highly efficient for $SO_2$ removal. The fluorine content of the circulating liquor may vary considerably according to the fluorine content of the coal being used. Fortunately, the fluorine problem can be eliminated with the use of a prescrubber (Type IV of Figure 3-2).

A venturi scrubber is efficient for dust removal but produces a high pressure drop. It may be preferable to use a combination of a highly efficient electrostatic precipitator (or baghouse) with a spray tower or grid-packed tower absorber which causes less pressure drop and a higher $SO_2$ removal efficiency.

## 3.4 FGD UNITS AT TAKEHARA

### 3.4.1 Overview

The Takehara power station has three boilers: No. 1, a coal-fired boiler (250 MW), No. 2 an oil-fired boiler (350 MW) and the new No. 3 coal-fired boiler currently under construction (700 MW). The FGD unit for the No. 1 boiler was constructed in 1977 by Babcock Hitachi Ltd.; the unit for the No. 3 boiler is under construction by Ishikawakima-Harima Heavy Industries. Both units use the limestone-gypsum process. The regulations which apply to these boilers are shown in Table 3-12. A description of the boiler, ESP, and FGD unit for the No. 1 boiler is given in Table 3-13.

EPDC also has constructed a demonstration plant for SCR of $NO_x$ in the flue gas from the No. 1 boiler. The SCR units are located upstream of the FGD unit.

### 3.4.2 Boiler No. 1

The important equipment and a flowsheet of the No. 1 FGD unit are shown in Table 3-14 and Figure 3-6 respectively. The unit has two scrubber systems in parallel. Each system consists of venturi and perforated tray scrubbers originally developed by Babcock and Wilcox, USA. Each system treats 426,000 $Nm^3$/hr of flue gas containing about 1,700 ppm $SO_2$ and 600 mg/$Nm^3$ of dust and removes about 90% of the $SO_2$ and dust. Limestone with 98% purity is used as the absorbent. The L/G ratio is 2.5 and 7 liters/$Nm^3$ for the venturi and tray scrubbers, while the pressure drop is 210 and 385 mm $H_2O$, respectively.

The slurry leaving the scrubber system is treated with sulfuric acid to lower the pH to about 4 and then subjected to air oxidation for conversion of the convert calcium sulfite to gypsum. Gypsum with 95% purity is produced at a rate of 12 tons/hr and is sold for wallboard and cement.

## TABLE 3-12. REGULATIONS WHICH APPLY TO BOILERS AT TAKEHARA STATION

| Air Pollutants | No. 1 Coal (250 MW) | No. 2 Oil (350 MW) | No. 3 Coal (700 MW) |
|---|---|---|---|
| Particulates, mg/Nm³ | 40 | 20 | 30 |
| Particulates, kg/hr | 117 | 117 | 117 |
| $SO_x$, Nm³/hr | 700 | 700 | 700 |
| $NO_x$, Nm³/hr | 394 | 394 | 394 |
| **Wastewater** | | | |
| pH | | 6.5 - 8.5 | |
| COD, ppm | | 10 | |
| SS, ppm | | 10 | |
| Oil, ppm | | 1 | |
| **Noise at Boundary Limit** | | | |
| 6:00 am - 10:00 pm, phon | | 60 | |
| Midnight, phon | | 50 | |

## TABLE 3-13. PLANT SPECIFICATIONS FOR TAKEHARA BOILER NO. 1

| | |
|---|---|
| **Boiler** | |
| Type | Opposite Firing (250 MW) |
| Supplier | Babcock Hitachi |
| Fuel | Coal (2% Sulfur) |
| **Dust Collector** | |
| Type | Cold-Side ESP |
| Particulate Outlet | Below 400 mg/Nm³ |
| **Flue Gas Desulfurization System** | |
| Process | Limestone-Gypsum |
| Supplier | Babcock-Hitachi |
| Gas Volume, Nm³/hr | 850,000 |
| $SO_2$ Removal, % | 94.2 (design, before reheating) |
| Dust Removal, % | 91.7 (design, before reheating) |
| Absorbent | Limestone 98% Purity |
| By-Product, t/hr | 12, Gypsum 95% purity |
| Gas Reheating | Oil-Firing |
| Water Supply, t/hr | 72 |
| Wastewater, t/hr | 12 |
| Commercial Start-Up | February 1977 |

TABLE 3-14. FGD EQUIPMENT FOR TAKEHARA BOILER NO. 1

| | |
|---|---|
| First Scrubber | |
| Number | 2 |
| Type | Venturi |
| Material | Acid Resistant Cast, Plastic-Lined |
| Gas Flow, $Nm^3$/hr | 426,000 each |
| Inlet $SO_2$, ppm | 1730 |
| Inlet Particulate, mg/$Nm^3$ | 600 |
| L/G Ratio, 1/$Nm^3$ | 2.5 |
| Pressure Drop, mm $H_2O$ | 230 |
| Second Scrubber | |
| Number | 2 |
| Type | Tray Tower with 5 Stages of Tray |
| Material | SS, Plastic Lined |
| L/G Ratio, 1/$Nm^3$ | 7 |
| Pressure Drop, mm $H_2O$ | 385 |
| Outlet $SO_2$, ppm | 100 (below 190 ppm after reheating) |
| Outlet Particulate, mg/$Nm^3$ | 40 |
| Mist Eliminator | |
| Type | Tube with Fin |
| Pressure Drop, mm $H_2O$ | 10 |
| Wash Rate, t/hr | 5.1 with Fresh Water |
| Thickener | |
| Number | 1 |
| Dimension, m | 14 (diameter) |
| Centrifuge | |
| Number | 9 (one is spare) |
| Inlet Solids, % | 20 |
| Outlet Solids, % | Over 88 |
| Fan, kW | 2900 x 2 |
| Pumps | |
| For First Scrubber | 2 (one is spare) x 150 kW |
| For Second Scrubber | 4 (one is spare) x 200 kW |

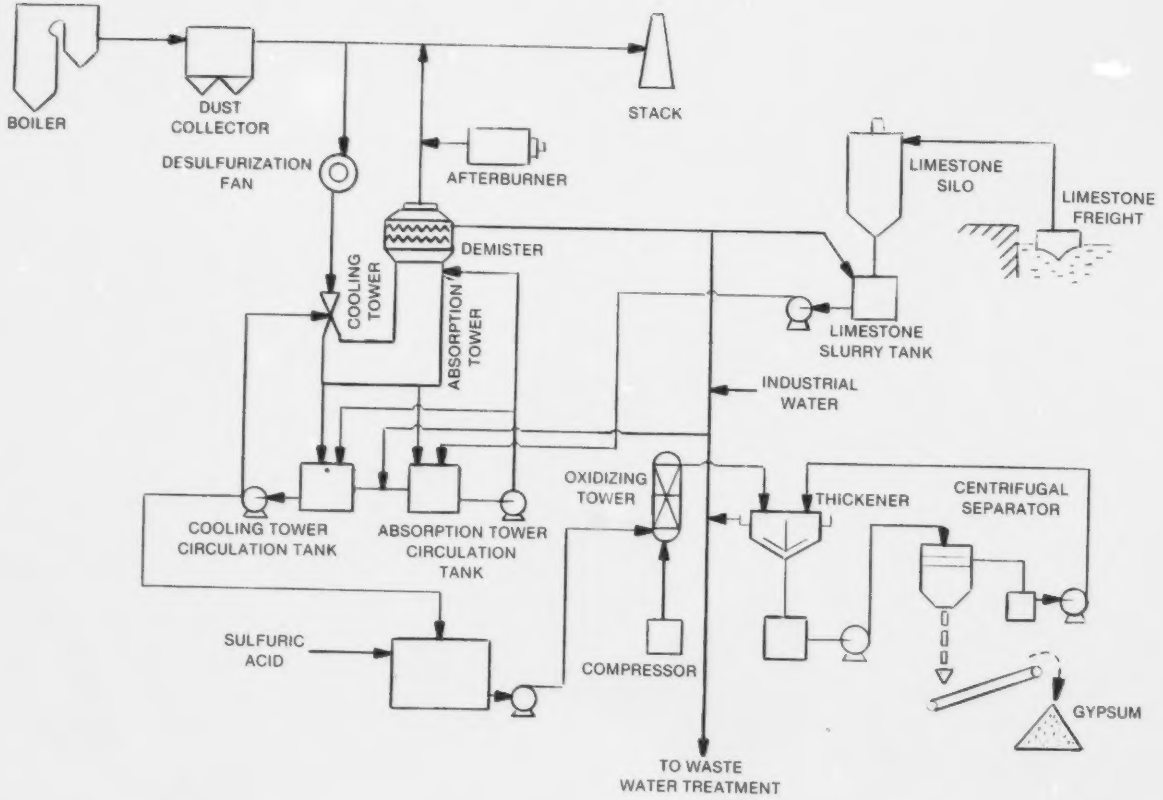
BOILER
DUST COLLECTOR
STACK
DESULFURIZATION FAN
AFTERBURNER
LIMESTONE SILO
LIMESTONE FREIGHT
DEMISTER
COOLING TOWER
ABSORPTION TOWER
LIMESTONE SLURRY TANK
INDUSTRIAL WATER
OXIDIZING TOWER
THICKENER
CENTRIFUGAL SEPARATOR
COOLING TOWER CIRCULATION TANK
ABSORPTION TOWER CIRCULATION TANK
SULFURIC ACID
COMPRESSOR
GYPSUM
TO WASTE WATER TREATMENT

Figure 3-6 Flowsheet of FGD Unit No. 1, Takehara.

In order to maintain a chloride concentration below 3,500 ppm in the circulating liquor, 12 tons/hr of wastewater are purged after being treated by the standard process described in Section 2.4.3 to meet the applicable regulations.

The low pressure drop mist eliminators consist of many tubes with fins and are washed primarily with fresh water.

The flue gas leaving the scrubber system is reheated by oil firing. A small amount of $SO_2$ is added to the flue gas when it is reheated but the overall $SO_2$ removal efficiency is maintained above 90% as shown in Table 3-15.

The Takehara Boiler No. 1 FGD unit has been operated with nearly 100% reliability since startup, as shown in Tables 3-2, 3-3 and 3-15. The unit has had some minor problems, including the plugging of the strainer in the slurry circulation tank. During the annual maintenance of the boiler, the strainer is removed for easy cleaning. Another problem has been scaling in the duct at the scrubber outlet due to mists. Presumably this is caused by the inefficiency of the low pressure drop mist eliminator.

The total investment for the FGD unit was 7.2 billion yen in 1977. The unit is operated by two persons per shift plus three persons in the daytime for maintenance. Each month it consumes 3,200 - 3,900 tons of limestone ground so that over 95% of it passes through a 325 mesh screen, 70-107 kiloliters of 98% sulfuric acid, and 40,000 - 46,000 tons of water. The unit produces 6,140 - 6,860 tons of byproduct gypsum. Power consumption accounts for 2.6% of the power generated by the boiler.

### 3.4.3 Demonstration Test of SCR-FGD Combination (Unit No. 1)

EPDC has begun demonstration tests using SCR of $NO_x$ and FGD on the flue gas from the No. 1 boiler at Takehara. As shown in Figure 3-7, the flue gas is treated by the two trains of a hot-side ESP, SCR reactor, air preheater, ID fan, and FGD system. One of the SCR reactors is manufactured

TABLE 3-15. PERFORMANCE OF FGD UNIT AT TAKEHARA (No. 1)

| | Hours | | | | Utilization (%) | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Month | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | Operability (C/B, %) | Reliability (C/D, %) | $SO_2$ Removal (%) |
| March 1980<br>April<br>May | 2208 | 2208 | 2208 | 2208 | 100 | 100 | 100 | 100 | 91 |
| June | 720 | 633[a] | 633[a] | 720 | 87.9 | 87.0 | 100 | 87.9 | 90.1 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 91.0 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 91.7 |
| September | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 93.1 |
| October | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.4 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 93.7 |
| December | 744 | 206[b] | 206 | 206 | 27.7 | 27.7 | 100 | 100 | 91.7 |
| January 1981 | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 92.5 |
| February | 672 | 672 | 670 | 672 | 100 | 100 | 100 | 100 | 91.5 |
| March | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 91.6 |
| April | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 91.8 |
| May | 744 | 168[c] | 168[c] | 168 | 22.6 | 22.6 | 100 | 100 | 92.9 |
| June | 720 | 110[c] | 110[c] | 110 | 15.2 | 15.2 | 100 | 100 | 94.3 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 94.1 |
| August | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.6 |
| September | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 94.4 |

[a] Boiler and FGD were shut down to remove scale at scrubber outlet.

[b] Boiler was shut down for scheduled maintenance.

[c] Boiler was shut down for annual maintenance.

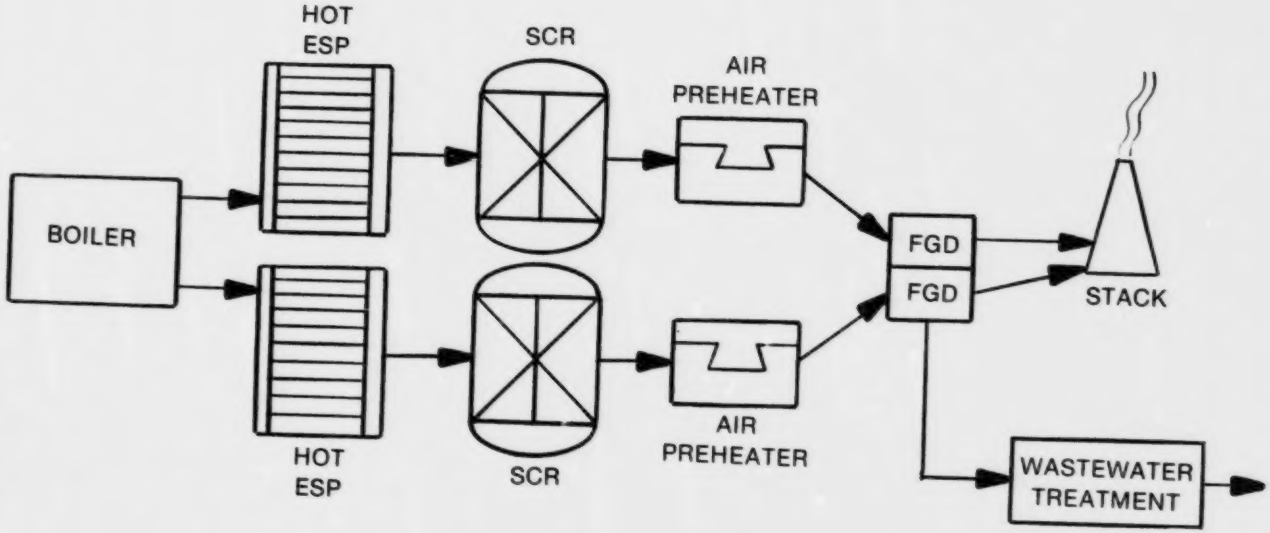
HOT
ESP
SCR
AIR
PREHEATER
BOILER
FGD
FGD
STACK
HOT
ESP
SCR
AIR
PREHEATER
WASTEWATER
TREATMENT

Figure 3-7 Demonstration Plant, Takehara Boiler No. 1.

by Babcock Hitachi Ltd., using a plate catalyst similar to that used at Hokkaido Electric's Tomato-Atsuma Station (Section 4.3), while the other is manufactured by Kawasaki Heavy Industries using a tubular catalyst.

A small amount of ammonia (less than 10 ppm) is contained in the flue gas leaving the SCR reactor and is caught by the scrubber. Since the Takehara Station faces the Seto Inland Sea which occasionally has a red tide problem, the ammonia concentration of the wastewater purged from the FGD unit must be kept below 10 ppm. To meet this requirement, Hitachi Zosen has constructed a conventional activated sludge treatment unit.

The SCR units were designed to remove over 80% of $NO_x$ in flue gas, and have been in operation since July 1981. During the initial tests a small amount of ammonium bisulfate deposited in the air preheater; this was removed by water washing. The deposition problem has almost been solved by reducing unreacted ammonia to below 5 ppm and by improved soot blowing.

### 3.4.4 FGD Unit for Takehara Boiler No. 3

A new coal-fired boiler No. 3 with a capacity of 700 MW was scheduled to begin operation at the Takehara Station in 1982. An FGD unit which uses a process similar to the one used at the Matsushima station is being built for the boiler by IHI. However, a major difference in the FGD systems is that at Takehara the flue gas is treated with two spray tower scrubbers in series. The sizes of spray tower scrubbers with different capacities are compared in Figure 3-8.

Another difference between this FGD system and that at the Matsushima station is the location of the fan. At Matsushima, the fan is placed at the gas-gas heater inlet at 140°C, while at Takehara No. 3 it is located between the heater and the FGD unit at 90-100°C (see Figure 2-8). The latter situation increases $SO_2$ removal efficiency; however, the fan must be made of a corrosion resistant material since sulfuric acid mists may condense at the lower temperatures. The No. 3 boiler has Sumitomo Heavy Industries hot ESPs for 99.5% dust removal and Babcock-Hitachi SCR reactors for 80% $NO_x$ removal.

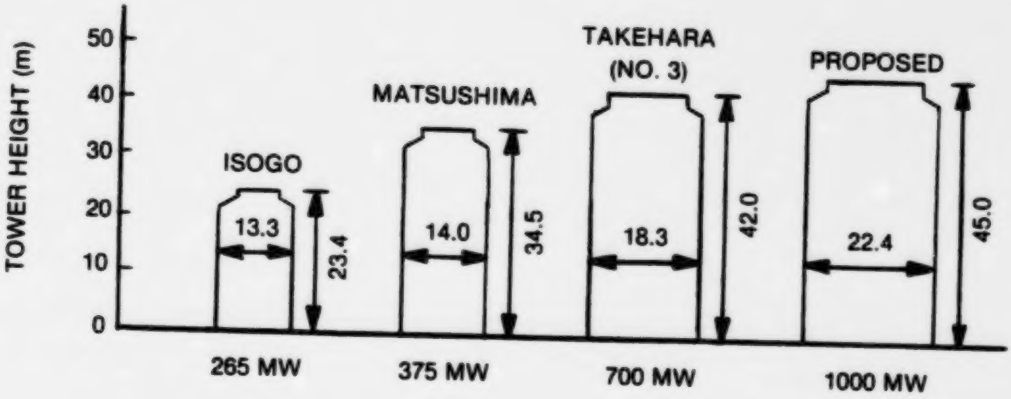

Figure 3-8 Comparison of Spray Tower Scrubbers with Varying Capacities.

### 3.4.5 Evaluation

The No. 1 FGD unit at Takehara has achieved a high $SO_2$ removal efficiency and reliability. The unit's major problem has been its high power consumption due to the large pressure drop. It may be preferable to use a low pressure drop reactor together with a highly efficient ESP.

The No. 3 FGD unit (as well as the Shimonoseki unit (Section 4.2), Tomato-Atsuma unit (Section 4.3) and Matsushima unit (Section 3.5)), has the above-mentioned improved features, and is expected to operate more successfully. The location of the fan between the gas-gas heater and the scrubber is desirable to attain a high $SO_2$ removal efficiency but may cause corrosion and dust deposit.

IHI is now designing a large spray scrubber which has the capacity to treat all the flue gas from a 1,000 MW coal-fired boiler. For actual use, two 500 MW scrubbers in parallel may be preferred to a single scrubber because most utility boilers only require one scrubber to operate at a reduced load (30-60% of full load) at night and on the weekend. It is expected that the air preheater downstream of the SCR reactor for the No. 3 boiler can be operated for about a year without a water wash. This is because both $SO_x$ and $NO_x$ concentrations in gas from the No. 3 boiler are lower than those from the No. 1 boiler; this causes lower $SO_3$ and $NH_3$ concentrations at the preheater inlet.

## 3.5 FGD UNITS AT MATSUSHIMA

### 3.5.1 Overview

EPDC recently constructed a new power station with two 500 MW coal-fired boilers at Matsushima, Kyushu. The boilers are fueled by imported coals containing about 1% sulfur. A limestone-gypsum process FGD unit with a capacity of treating three-fourths of the flue gas was constructed by IHI for boiler No. 1; a similar unit was built by Babcock-Hitachi Ltd. for boiler No. 2 (Figure 3-9). The former went into operation in late 1980, the latter in October 1981.

Table 3-16 gives a description of the regulations, boiler, and the ESP and FGD systems for the No. 1 system. The No. 2 system is similar to the No. 1 system. A flowsheet of the system is shown in Figure 3-10. The flue gas from the boiler contains about 1,000 ppm $SO_2$ and 20 $g/Nm^3$ of dust with a flow rate of 1,651,000 $Nm^3/hr$. It is first treated by a hot-side ESP to remove 99.3% of the dust. The hot-side ESP was chosen because the boiler will burn various imported coals including low-sulfur ones. The gas is then cooled to 135-150°C in an air preheater. Three-fourths of the gas is passed through two Ljungstrom gas-gas heaters in parallel where it is cooled to 80-85°C. The gas then passes through the FGD system which removes over 90% of the $SO_2$.

The FGD units have a prescrubber to remove flue gas impurities such as particulates, chloride, and fluoride. Prescrubbing protects the scrubber system from erosion and corrosion, increases $SO_2$ removal efficiency, prevents scaling, and increases the purity of byproduct gypsum. About 10.5 t/hr per unit of 98% pure gypsum salable for cement and wallboard is produced.

### 3.5.2 FGD Unit No. 1

The specifications of the prescrubber and scrubber of unit No. 1 are given in Table 3-17. The prescrubber is a Chemico spray tower. About 80% of the particulates and virtually all of the fluoride and chloride in the flue gas which has a L/G ratio of 3.0 $l/Nm^3$ are removed in the prescrubber. The pressure drop across the prescrubber is 45 mm $H_2O$.

The flue gas leaving the prescrubber at about 45°C is introduced into a Chemico spray tower scrubber through a mist eliminator. The scrubber has 6 stages of spray nozzles which remove about 94% of the $SO_2$ using a limestone

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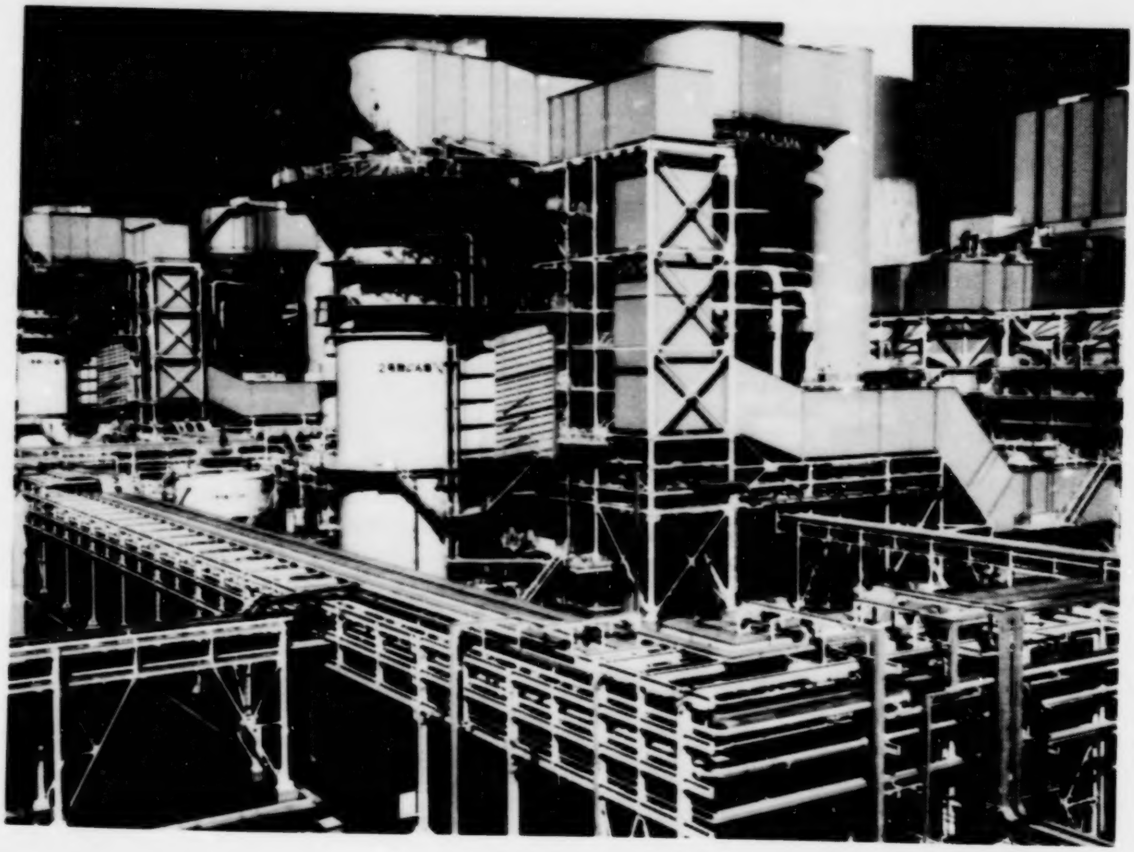

Figure 3-9 Matsushima Plant, EPDC (No. 2 Unit at Center, No. 1 Unit Left)

TABLE 3-16. DESCRIPTION OF MATSUSHIMA NO. 1 UNIT

| | |
|---|---|
| **Regulations** | |
| Particulate, mg/Nm$^3$ | 100 |
| SO$_2$, Nm$^3$/hr | 402 (260 ppm) |
| **Boiler** | |
| Capacity, MW | 500 |
| Type | Corner Firing |
| Supplier | Mitsubishi Heavy Industries |
| Service Load | Base Load |
| Flue Gas Volume, Nm$^3$/hr | 1,651,000 |
| Sulfur in Coal, % | 1.2 (design) |
| **Electrostatic Precipitator (Hot Side)** | |
| Supplier | Sumitomo Heavy Industries |
| Number | 1 |
| Particulate Removal Efficiency, % | 99.3 |
| Particulate Outlet, mg/Nm$^3$ | 100 |
| **FGD** | |
| Gas Treated, Nm$^3$/hr | 1,300,000 (79% of total) |
| Process | Limestone-Gypsum |
| Supplier | Ishikawajima-Harima Heavy Industries (IHI) |
| Inlet SO$_2$, ppm | 1000 |
| SO$_2$ Removal Efficiency, % | 95 (design) |
| Limestone, t/hr | 5.5 (98% Purity)<br>(95% under 325 Mesh) |
| By-product Gypsum, t/hr | 10.5 (over 98% Purity) |
| Water Consumption, t/hr | 50 |
| Wastewater, t/hr | 15 |

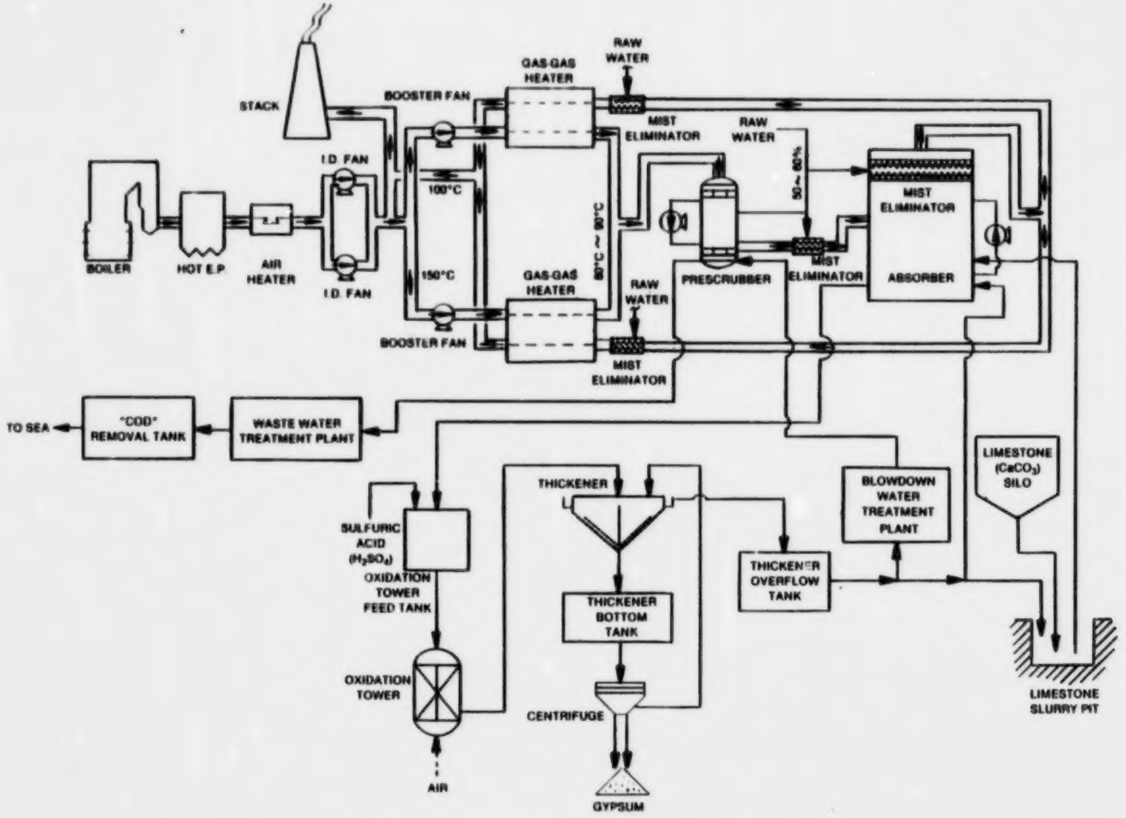
STACK
BOOSTER FAN
GAS-GAS HEATER
RAW WATER
MIST ELIMINATOR
RAW WATER
I.D. FAN
100°C
50 ~ 60%
MIST ELIMINATOR
BOILER
HOT E.P.
AIR HEATER
I.D. FAN
150°C
80°C ~ 90°C
GAS-GAS HEATER
RAW WATER
PRESCRUBBER
MIST ELIMINATOR
ABSORBER
BOOSTER FAN
MIST ELIMINATOR
TO SEA
"COD" REMOVAL TANK
WASTE WATER TREATMENT PLANT
LIMESTONE (CaCO₃) SILO
THICKENER
BLOWDOWN WATER TREATMENT PLANT
SULFURIC ACID (H₂SO₄)
OXIDATION TOWER FEED TANK
THICKENER OVERFLOW TANK
THICKENER BOTTOM TANK
LIMESTONE SLURRY PIT
OXIDATION TOWER
CENTRIFUGE
AIR
GYPSUM

FIGURE 3-10 Flowsheet for FGD Unit, EPDC Matsushima Station.

## TABLE 3-17. SCRUBBER SPECIFICATIONS (MATSUSHIMA NO. 1)

| | |
|---|---|
| **Prescrubber (Gas Cooling and Dust Removal)** | |
| Number | 1 (No standby) |
| Type | Spray Tower (Chemico type) |
| Dimensions | 9.4 m (Diameter), 32 m (height) |
| Shell Material | Mild Steel |
| Shell Lining Material | Flake Glass Filled Plastic |
| Gas Treated, $Nm^3/hr$ | 1,300,000 |
| Inlet Temperature, °C | 110 (max.) |
| Outlet Temperature, °C | 45 |
| Liquid Recirculation Rate, $m^3/hr$ | 4,260 |
| L/G Ratio, $1/m^3$ | 3.0 |
| Particulate Concentration, $mg/Nm^3$ | |
| (Inlet) | 300 (design) |
| (Outlet) | 50 (design) |
| Particulate Removal Efficiency, % | 83.3 |
| $SO_2$ Inlet Load, $Nm^3/hr$ | 1,215 |
| $SO_2$ Removal | None |
| Pressure Drop, mm $H_2O$ | 45 |
| **Scrubber ($SO_2$ Removal)** | |
| Number | 1 (no standby) |
| Type | Spray Tower (Chemico type) |
| Dimensions | 14.0 m (diameter), 34.5 m (height) |
| Shell Material | Mild Steel |
| Shell Lining Material | Flake Lining |
| Gas Flow, $Nm^3/hr$ | 1,300,000 |
| Gas Temperature, °C | 45 |
| Liquid Recirculation Rate, $m^3/hr$ | 20,400 |
| L/G Ratio, $1/Nm^3$ | 15.7 |
| Superficial Gas Velocity, m/sec | 3.0 |
| Particulate Inlet Concentration, $mg/Nm^3$ | 50 |
| Particulate Outlet Concentration, $mg/Nm^3$ | 30 |
| Particulate Removal, % | 40 |
| $SO_2$ Inlet Load, $Nm^3/hr$ | 1,215 |
| $SO_2$ Outlet Load, $Nm^3/hr$ | 73 |
| $SO_2$ Inlet Concentration, ppm | 935 |
| $SO_2$ Outlet Concentration, ppm | 55 |
| Removal Efficiency, % | 94 |
| Pressure Drop, mm $H_2O$ | 100 |

slurry with an L/G ratio of 15.5 $l/Nm^3$. The limestone has 98% purity and is ground finely so that 95% of it can pass through a 325 mesh screen. The pressure drop across the scrubber is 100 mm $H_2O$. About 40% of the dust in the flue gas is also removed in the scrubber. Thus the gas leaving the scrubber contains less than approximately 55 ppm $SO_2$ and less than 30 $mg/Nm^3$ of dust. The relationship between the L/G ratio and $SO_2$ removal efficiency in the spray tower is shown in Figure 3-11. The gas leaving the scrubber passes through a mist eliminator and then goes to the gas-gas heater where it is heated to 100-110°C. Since a small portion of the gas leaks into the heater, the overall $SO_2$ removal efficiency is about 90%.

The calcium sulfite slurry leaving the scrubber is treated with sulfuric acid to lower its pH to about 4 and then treated with air in the oxidizer to convert the sulfite to gypsum. The 18% concentrated gypsum slurry is thickened to about 20% concentration and then centrifuged to yield byproduct gypsum containing 7-9% moisture. The byproduct gypsum has a purity of over 95% and is sold for cement and wallboard. The specifications of various pieces of equipment in the FGD unit are shown in Table 3-18.

The mist eliminators are washed with fresh water to prevent scaling. Wastewater is blown down from the scrubber system and treated with the wastewater from the prescrubber at a rate of 15 t/hr using the standard process described in Section 2.4.3. In order to reduce the amount of wastewater, the FGD unit has a facility which treats the blowdown water from the scrubber system with sodium carbonate and separates the precipitated calcium carbonate by filtration. This enables the softened water to be fed to the prescrubber for gas cooling. By using this facility, both the wastewater and fresh water requirements can be reduced although sodium carbonate is consumed.

As shown in Table 3-19, the unit has operated with 100% reliability since startup in January 1981. A major problem has been dust deposition in the gas heater. The pressure drop across the heater was initially 40 mm $H_2O$ and increased to 140 mm $H_2O$ after one year of operation. An inspection

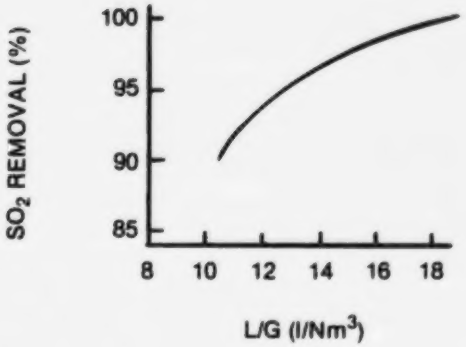

**Figure 3-11** **L/G vs. $SO_2$ Removal Efficiency (12).**

(Spray tower, pH = 5.6; inlet $SO_2$ = 800 ppm)

70A2500

### TABLE 3-18. SPECIFICATIONS OF EQUIPMENT AT MATSUSHIMA NO. 1

| | |
|---|---|
| **Mist Eliminator** | |
| Type | Chevron (horizontal flow) |
| Wash | Continuous |
| Wash Liquor | Fresh Water and Gypsum Filtrate |
| **Reheater** | |
| Type | Gas-Gas Heater (Ljungstrom) |
| Gas Temperature | 100°C After Reheating |
| Number | 2 |
| **Thickener** | |
| Inlet Solid | 7-9% |
| Outlet Solid | 20.0% |
| **Centrifuge** | |
| Type | Basket |
| Material | Stainless Steel |
| Number | 7 (1 for standby) |
| Inlet Solids | 20% |
| Outlet Solids | 90% |
| **Pumps** | |
| For Scrubbers | 240 kW x 12 (2 for standby) |
| For Oxidizer Feed | 15 kW x 2 (1 for standby) |
| For Centrifuge feed | 22 kW x 1 (1 for standby) |
| **Fan** | 3500 kW x 2 |

## TABLE 3-19. PERFORMANCE OF FGD UNIT AT MATSUSHIMA (No. 1)

| Month | Hours | | | | Utilization (%) | | Oper- ability (C/B, %) | Reli- ability (C/D, %) |
|---|---|---|---|---|---|---|---|---|
| | Total (A) | Boiler oper- ation (B) | FGD oper- ation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | | |
| January 1981 | 744 | 368 | 368 | 368 | 49.5 | 49.5 | 100 | 100 |
| February | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 |
| March | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| April | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| May | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| June | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| August | 744 | 713.6 | 713.6 | 713.6 | 95.9 | 95.9 | 100 | 100 |
| September | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| October | 744 | 711.1 | 711.1 | 711.1 | 95.6 | 95.6 | 100 | 100 |
| November | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| December | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| Total | 8760 | 8158.7 | 8158.7 | 8158.7 | 93.1 | 93.1 | 100 | 100 |

during the annual boiler shutdown period revealed the presence of deposits in the low temperature zone. Efforts have since been made to reduce the deposits. The unit consumes 8,300 kW of power, 5.5 t/hr of limestone and 0.26 t/hr of sulfuric acid. It also produces 10.5 t/hr of gypsum. The No. 1 and No. 2 FGD units together are operated by two persons per shift; the operation is almost fully-automated. In addition, during the day several people handle the limestone and gypsum and perform maintenance.

### 3.5.3 Evaluation

The operation of the No. 1 FGD unit at Matsushima has been carried out successfully and the No. 2 unit has successfully begun operation as well. Since FGD technology is well developed, it is not unusual for a limestone scrubbing unit without a stand-by scrubber to startup and operate at 100% reliability. Normally there is a test operation period for a few months prior to the startup when adjustments are made.

The Matsushima FGD system is similar to those of Chugoku Electric at Shimonoseki constructed by MHI (Section 4.2) and of Hokkaido Electric at Tomato-Atsuma constructed by Babcock Hitachi (Section 4.3) in that it uses a gas-gas heater, prescrubber, low pressure drop scrubber, and sulfuric acid addition for pH control. The softening unit for the scrubber liquor also is used at the Tomato-Atsuma unit.

The Matsushima units use spray tower absorbers while the Shimonoseki unit uses a plastic grid-packed tower and the Tomato-Atsuma unit uses a perforated plate scrubber. Long-term operation will reveal the advantages and disadvantages of the scrubber types.

At the present time such a system is probably the most advanced type of limestone-gypsum FGD process available for coal-fired boilers to attain a high $SO_2$ removal efficiency and reliability with minimum water consumption and wastewater.

# SECTION 4

# OTHER COMMERCIAL FGD UNITS IN OPERATION FOR COAL-FIRED BOILERS

## 4.1 OMUTA PLANT, MITSUI ALUMINUM

### 4.1.1 History

The Mitsui Aluminum Omuta Plant has two coal-fired boilers, 156 MW and 175 MW, which burn domestic coal containing about 2% sulfur. The boilers are used for power generation for the aluminum plant and have operated normally at constant loads.

The FGD unit for the No. 1 boiler (156 MW) was constructed by Mitsui Miike Machinery Co. and went into operation in 1972 as the first FGD unit for a coal-fired boiler in Japan. The unit uses Chemico two-stage venturi scrubbers and is the only large-scale unit in Japan to produce throw-away sludge. Carbide lime is used as the absorbent. The unit has two identical size scrubbers in parallel, one of which was originally used for standby purposes.

Initially, about 70% of the 512,000 $Nm^3/hr$ flue gas produced was treated to meet the regulation with a k value of 11.7. An explanation of the k value can be found in Section 2 of this report. In 1976, when the k value was reduced to 4.03, both scrubbers were put into operation and treated the entire gas stream at an 89% $SO_2$ removal efficiency. The k value was further reduced to 3.33 in 1977 when additional slurry recirculation pumps were installed. These pumps increased the L/G ratio from 8-9 $l/Nm^3$ to 11-12 $l/Nm^3$ achieving over 90% $SO_2$ removal efficiencies. In 1978, the total emission regulation was applied to limit $SO_x$ emissions from the No. 1 boiler to a level below 109.3 $Nm^3/hr$ (k value 3.06, $SO_2$ below 213 ppm).

Regulations were further tightened in 1980 to maintain $SO_x$ emissions below 106.7 Nm3/hr (k value 2.99, $SO_x$ below 208 ppm, over 91% $SO_x$ removal). The new regulations were met by slightly raising the pH of the absorbent slurry to about 7.5. The operation of the FGD unit has been very successful, achieving virtually 100% reliability since startup.

The No. 2 boiler and its FGD unit began operation in 1975 to meet the regulation with a k value of 2.71. The boiler has a capacity of 175 MW and produces a flue gas flow of 552,000 $Nm^3$/hr. Since both carbide lime use and sludge pond facilities are not sufficient to accommodate both FGD units, a limestone-gypsum process developed by Mitsui Miike Machinery Co. has been used for the No. 2 FGD unit. The unit uses two Chemico venturi scrubbers in series.

In 1977, a spray tower scrubber was added to treat 150,000 $Nm^3$/hr of the flue gas, leaving the existing scrubbers to treat the remaining 400,000 $Nm^3$/hr. The addition of this scrubber increased the $SO_x$ removal efficiency to about 90%, and enabled the boiler to meet the stricter requirements of the new regulation with the k value of 2.13. In 1978, the total emission regulation applied to the No. 2 boiler restricted $SO_x$ emissions to below 117.5 $Nm^3$/hr (k value 1.97). In 1981, a Ljungstrom gas-gas heater was installed so that gas, rather than fuel oil, could be used for reheating purposes. The No. 2 FGD unit has also operated successfully, removing over 90% of the $SO_x$ in the flue gas and producing salable gypsum.

### 4.1.2 No. 1 FGD Unit

A flowsheet of the No. 1 FGD unit is shown in Figure 4-1. Figure 4-2 illustrates the scrubber and sludge pond. The flue gas has a flow rate of 512,000 $Nm^3$/hr and leaves the ESP at about 140°C containing 2,000 - 2,200 ppm $SO_2$ and 500 - 600 mg/$Nm^3$ of dust. The gas is introduced into 2 scrubbers in parallel, each of which has a two-stage venturi, and removes over 90% of the $SO_x$ and dust present in the stream. This efficiency is achieved by feeding an absorbing slurry at pH 7.2 - 7.8 containing carbide lime at an inlet stoichiometry of 1.05 into each scrubber. The pressure

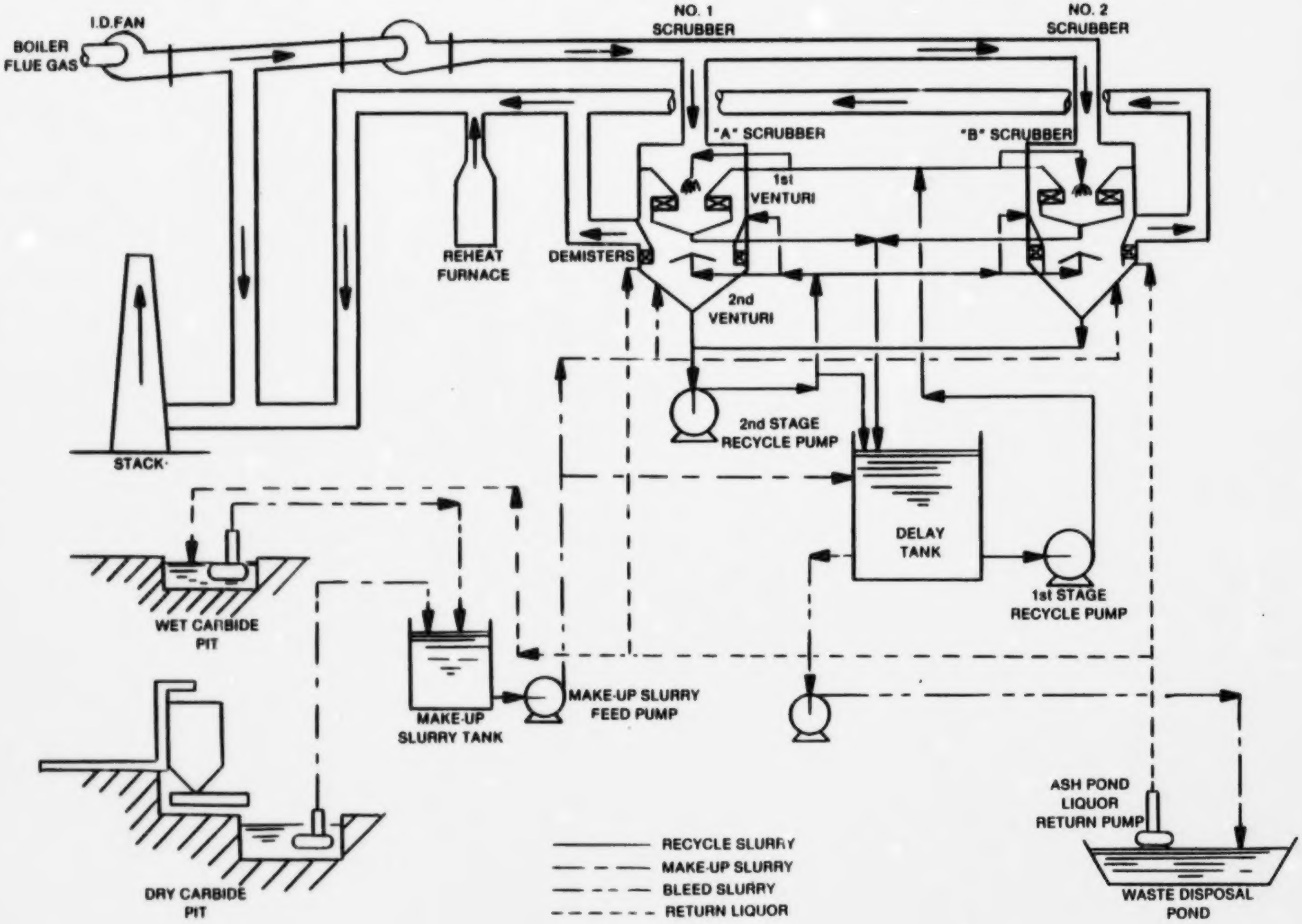
I.D.FAN
BOILER FLUE GAS
NO. 1 SCRUBBER
NO. 2 SCRUBBER
"A" SCRUBBER
"B" SCRUBBER
1st VENTURI
2nd VENTURI
REHEAT FURNACE
DEMISTERS
STACK
2nd STAGE RECYCLE PUMP
DELAY TANK
1st STAGE RECYCLE PUMP
WET CARBIDE PIT
MAKE-UP SLURRY TANK
MAKE-UP SLURRY FEED PUMP
DRY CARBIDE PIT
ASH POND LIQUOR RETURN PUMP
WASTE DISPOSAL POND
RECYCLE SLURRY
MAKE-UP SLURRY
BLEED SLURRY
RETURN LIQUOR

Figure 4-1 Flowsheet of the No. 1 FGD Unit, Mitsui Aluminum (Carbide Lime Adsorption, Throw-Away Sludge).

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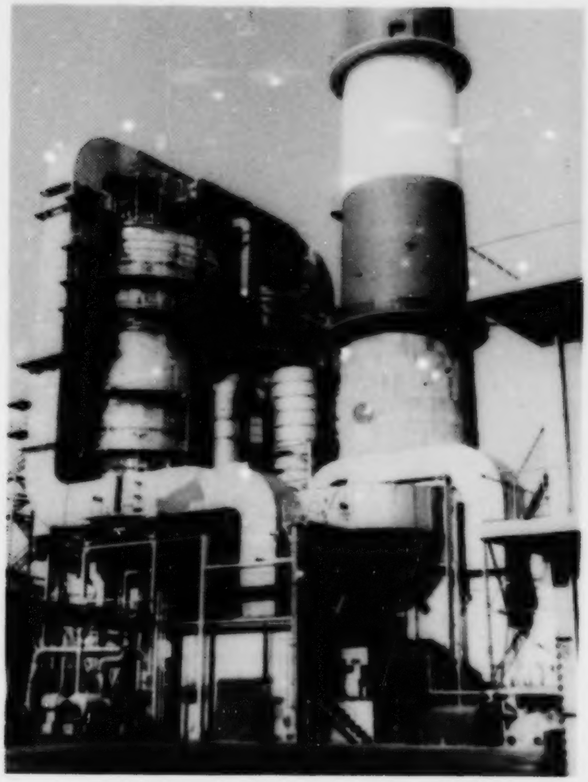


**Figure 4-2** Unit No. 1 FGD System of Mitsui Aluminum at Omuta (Scrubber and Sludge Pond)

drop through the scrubbers, including the Chevron mist eliminators, is 180 mm $H_2O$. The flue gas leaving the scrubber at 50°C is heated to 80°C by an oil-fired burner.

The slurry concentration in the FGD system is 6-8 percent. The L/G ratio in the scrubber is 8-9 $l/Nm^3$. Slurry containing calcium sulfite and fly ash with a small amount of impurities is sent to a waste disposal pond through a holding tank. The pond supernatant is recycled to the scrubber system. After the number of recirculation pumps was increased in 1977, the power requirement of the FGD system became 4,200 kW. This power consumption accounts for 2.7% of the power generated by the boiler.

The FGD system has operated successfully since its startup in 1972 with virtually 100% reliability. System performance during the past year is shown in Table 4-1. Although the FGD system has experienced minor problems such as small amounts of scale formation and pump erosion, the scrubber system can be operated continuously for one year until the boiler is shut down for annual maintenance. In this way, maintenance of the FGD system can coincide with scheduled boiler maintenance.

The total investment cost of the No. 1 FGD system was 1.03 billion yen in 1972.

### 4.1.3 No. 2 FGD Unit

A flowsheet of the No. 2 boiler FGD unit is shown in Figure 4-3. Flue gas leaving the ESP at 140°C containing 2,000-2,200 ppm $SO_x$ with 500-600 $mg/Nm^3$ of dust is passed through a Ljungstrom gas-gas heater where it is cooled to 100°C. A major portion of the gas is then introduced into two venturi scrubbers in series and treated with limestone slurry to remove over 90% of the $SO_x$ and dust in the stream. The slurry contains a catalyst which promotes $SO_2$ removal and oxidation of calcium sulfite to gypsum. The L/G ratio is 13-15 $l/Nm^3$ for the first scrubber and 9-11 $l/Nm^3$ for the second scrubber. The calcium slufite slurry leaves the first scrubber at a pH of about 5.5 and is sent to the pH controller where contact with flue

TABLE 4-1. PERFORMANCE OF NO. 1 FGD UNIT AT OMUTA, MITSUI ALUMINUM (156 MW)

|  | Hours |  |  |  | Utilization (%) |  |  |  |  |  |
|---|---|---|---|---|---|---|---|---|---|---|
| Month | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | Operability (C/B,%) | Reliability (C/D,%) | $SO_2$ Inlet (ppm) | $SO_2$ Removal[b] (%) |
| Sept. 1980 | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 2150 | 90.9 |
| Oct. | 744 | 5[a] | 0 | 0 | - | - | - | - | - | - |
| Nov. | 720 | 469[a] | 464 | 464 | 65.1 | 64.4 | 89.8 | 100 | 1840 | 89.3 |
| Dec. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2150 | 91.0 |
| Jan. 1981 | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2120 | 90.9 |
| Feb. | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 2150 | 91.0 |
| Mar. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2140 | 90.9 |
| Apr. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 2160 | 91.1 |
| May | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 100 | 2150 | 91.1 |
| June | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 2140 | 91.0 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2150 | 90.8 |
| Aug. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2120 | 90.9 |

[a] Boiler was shut down for annual maintenance. Maintenance of FGD unit was also made. Scale, formed in a small amount in the scrubber system, was removed.

[b] After reheating by oil-firing, outlet $SO_2$ is kept at 192 - 197 ppm.

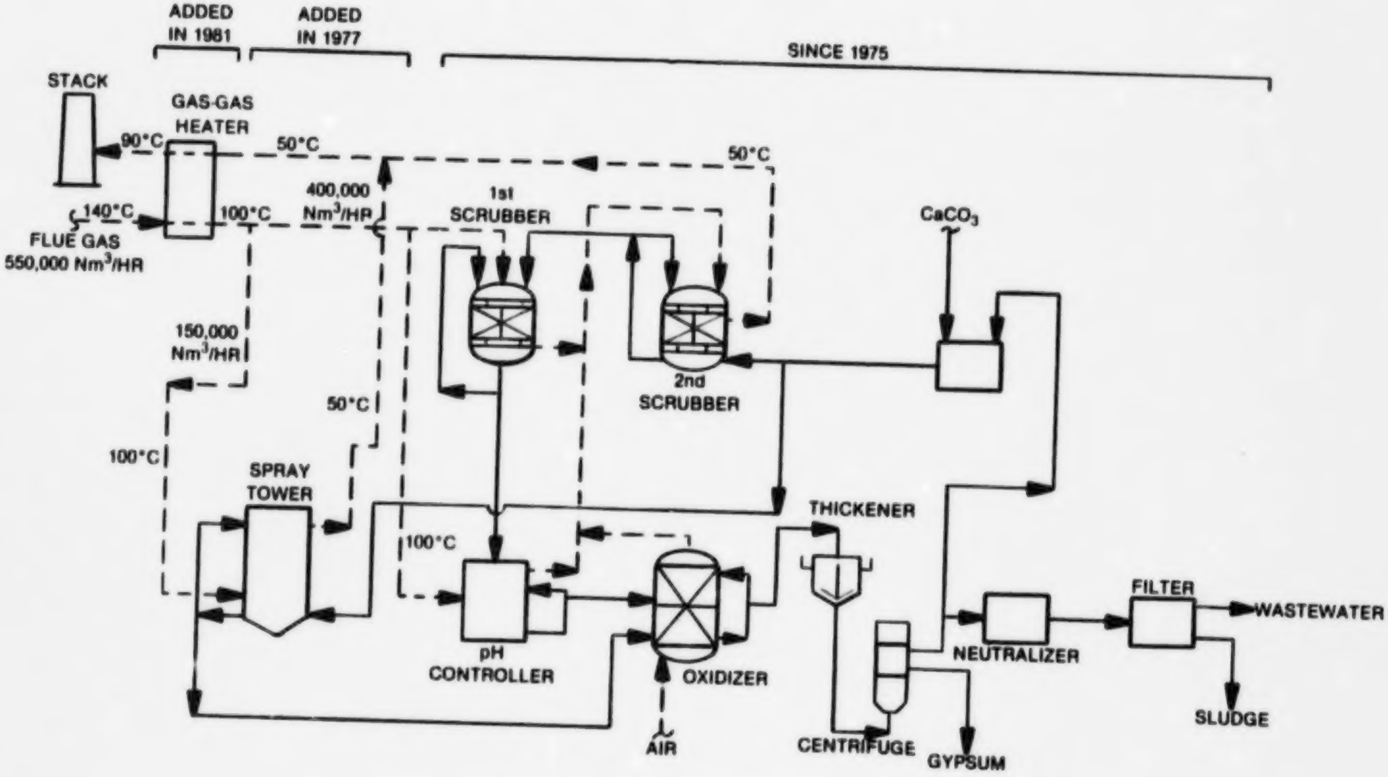

ADDED IN 1981
ADDED IN 1977
SINCE 1975
STACK
GAS-GAS HEATER
90°C
50°C
50°C
140°C
FLUE GAS 550,000 Nm³/HR
100°C
400,000 Nm³/HR
1st SCRUBBER
CaCO₃
150,000 Nm³/HR
2nd SCRUBBER
50°C
100°C
SPRAY TOWER
THICKENER
100°C
pH CONTROLLER
OXIDIZER
FILTER
WASTEWATER
NEUTRALIZER
SLUDGE
AIR
CENTRIFUGE
GYPSUM

Figure 4-3 Flowsheet of the No. 2 FGD Unit, Mitsui Aluminum*

70A2502

*Spray Tower and Gas-Gas Heater were Added Later.

gases lowers the pH to about 5. The slurry is then oxidized by air to form gypsum; the gypsum is treated by a thickener and centrifuged into a salable form. The flue gas leaving the pH controller and the air leaving the oxidizer contain small amounts of $SO_2$ and are sent to the second scrubber. The flue gas leaving the second scrubber is heated in the gas-gas heater to 90°C. This FGD system is similar to EPDC's units at the Takasago Station (Section 3.2).

Before 1977, all of the flue gas was treated by the venturi scrubber system for removal of nearly 90% of the $SO_x$. In 1977, a spray tower absorber was added to treat 150,000 $Nm^3$/hr of flue gas, thereby increasing the $SO_x$ removal efficiency to above 90%. A limestone slurry is used in the scrubber. The calcium sulfite slurry leaves the absorber at a pH of about 4 and is sent to the oxidizer. Flue gas leaving the absorber is heated by the gas-gas heater together with the flue gas leaving the second venturi scrubber.

The No. 2 FGD unit has been operated successfully since startup, as shown in Table 4-2. The No. 2 unit consumes 6,000 kW of electric power, which is 3.5% of the power generated by the boiler. Twenty-five to 30 t/hr of wastewater are purged after being treated. Chloride concentrations in the scrubber liquor have been maintained at about 500 ppm. The proprietary catalyst is neither expensive nor hazardous to dispose of and is purged with the wastewater.

The total investment cost for the No. 2 system was 5.5 billion yen. Both No. 2 and No. 1 systems have been operated by 3 persons per shift. The annualized cost of desulfurization by the No. 2 unit is 1.8 yen/kWhr based on a 7 year depreciation schedule.

### 4.1.4 Evaluation

The FGD unit for the No. 1 boiler may be one of the most successful units in the world using a throw-away sludge system. Before the unit was

TABLE 4-2. PERFORMANCE OF NO. 2 FGD UNIT AT OMUTA, MITSUI ALUMINUM (175 MW)

| Month | Hours | | | | Utilization (%) | | Operability (C/B,%) | Reliability (C/D,%) | $SO_2$ Inlet (ppm) | $SO_2$ Removal[b] (%) |
|---|---|---|---|---|---|---|---|---|---|---|
| | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | | | | |
| Sept. 1980 | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 2160 | 91.2 |
| Oct. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2050 | 90.7 |
| Nov. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 1960 | 90.3 |
| Dec. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2080 | 90.9 |
| Jan. 1981 | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2160 | 91.2 |
| Feb. | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 2090 | 90.9 |
| Mar. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2120 | 91.1 |
| Apr. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 2130 | 91.1 |
| May | 744 | 131[a] | 126 | 126 | 17.6 | 16.9 | 96.1 | 100 | 2160 | 91.2 |
| June | 720 | 212[a] | 207 | 207 | 29.4 | 28.8 | 97.6 | 100 | 2020 | 90.6 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2090 | 90.9 |
| Aug. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 2100 | 90.9 |

[a] Boiler was shut down for annual maintenance; maintenance of the FGD system was conducted; a small amount of scale in the scrubber system was removed; the gas-gas heater was connected.

[b] After reheating by a gas-gas heater, outlet $SO_2$ is kept at 189-191 ppm.

installed, Mitsui Aluminum and Mitsui Miike Machinery Co. conducted extensive pilot plant tests using a Chemico venturi scrubber. At that time, they experienced many problems including serious scaling. The smooth operation of the commercial unit is due to its improved design, good fabrication, suitable operational control, and an unsaturated mode operation which prevents the formation of gypsum (Section 2.3.3). Microscope and x-ray tests conducted in 1981 after nine years of operation showed that the calcium sulfite sludge at the scrubber outlet did not contain gypsum. However, a small amount of sulfate was detected by chemical analysis. This indicates that the sulfate is contained in the crystal lattice of the calcium sulfite and does not accumulate in the scrubber liquor.

Although the throw-away system has been operated successfully in this case, it is not likely to be used in future large scale applications in Japan due to the limited amount of land available for disposal areas.

The No. 2 boiler FGD unit has also been operated successfully. The unit is characterized by the use of catalysts and pH controllers to lower the pH of the slurry without adding sulfuric acid. The No. 2 unit is similar to the Takasago units of EPDC (Section 3.2). This particular system, however, is more complex and requires considerable energy. If sulfuric acid is ever available at a low cost, pH control by sulfuric acid addition may be preferable to this energy-intensive system.

Venturi scrubbers are highly efficient for dust removal but not for $SO_2$ removal. By using more efficient scrubbers such as packed towers or spray towers, over 90% $SO_2$ removal can be attained using a limestone slurry without a catalyst. Venturi scrubbers also produce a large pressure drop resulting in high power consumption. Use of a highly efficient ESP and a low pressure drop scrubber such as a grid-packed tower or spray tower may be preferable in order to cut energy costs.

## 4.2 SHIMONOSEKI PLANT, CHUGOKU ELECTRIC

### 4.2.1 History

The Shimonoseki Station of the Chugoku Electric Power Co. has two boilers--a 175 MW coal-fired boiler (No. 1) and a 400 MW oil-fired boiler (No. 2). Regulations for the station are shown in Table 4-3.

TABLE 4-3. REGULATIONS FOR SHIMONOSEKI STATION

| Pollutant | Regulated Level |
|---|---|
| Air Pollution Control | |
| k Value | 2.7 (Ground level $SO_2$ concentration of 0.0047 ppm) |
| $SO_x$ (total) | Below 412 $Nm^3$/hr |
| Particulates | Below 130 kg/hr |
| No. 1 Boiler | Below 200 mg/$Nm^3$ |
| No. 2 Boiler | Below 40 mg/$Nm^3$ |
| $NO_x$ | Below 330 $Nm^3$/hr |
| No. 1 Boiler | Below 350 ppm |
| No. 2 Boiler | Below 170 ppm |
| Suspended particulates | Below 0.2 mg/$m^3$ |
| Water Pollution Control | |
| pH | 5.8 - 8.6 |
| Suspended Solids | Below 12 kg/day<br>Below 15 mg/liter |
| Normal-hexane-soluble material | Below 0.8 kg/day<br>Below 1 mg/liter |
| Chemical oxygen demand | Below 12 kg/day<br>Below 15 mg/liter |

The No. 1 boiler at Shimonoseki was completed in 1967 and burned coal and oil in a 25 to 75 ratio before a full scale MHI wet limestone-gypsum process FGD plant was completed in July 1979. After the FGD plant became operational, coal and oil were used in a 50/50 ratio. It was difficult to use larger amounts of coal because of the $NO_x$ regulation (below 350 ppm). Although the regulation could be met by combustion modification, even with the exclusive burning of coal, it was likely that an additional $NO_x$ reduction might be required in the future. Therefore, Chugoku Electric decided to install a full-scale SCR unit to allow for combustion of coal only. The SCR unit, completed in March 1980, is the first full-scale SCR demonstration plant for a coal-fired boiler in the world.

Figure 4-4 shows the combined SCR/FGD system for the No. 1 boiler. In the system the flue gas is first subjected to SCR at 330-400°C, then passed through 2 trains of air preheaters and dust collectors (multicyclone and ESP), and finally undergoes FGD after it has passed through a heat exchanger. A description of the boiler and the flue gas treatment system is given in Table 4-4.

The relatively new No. 2 boiler uses a high-sulfur oil with MHI wet limestone-gypsum process FGD.

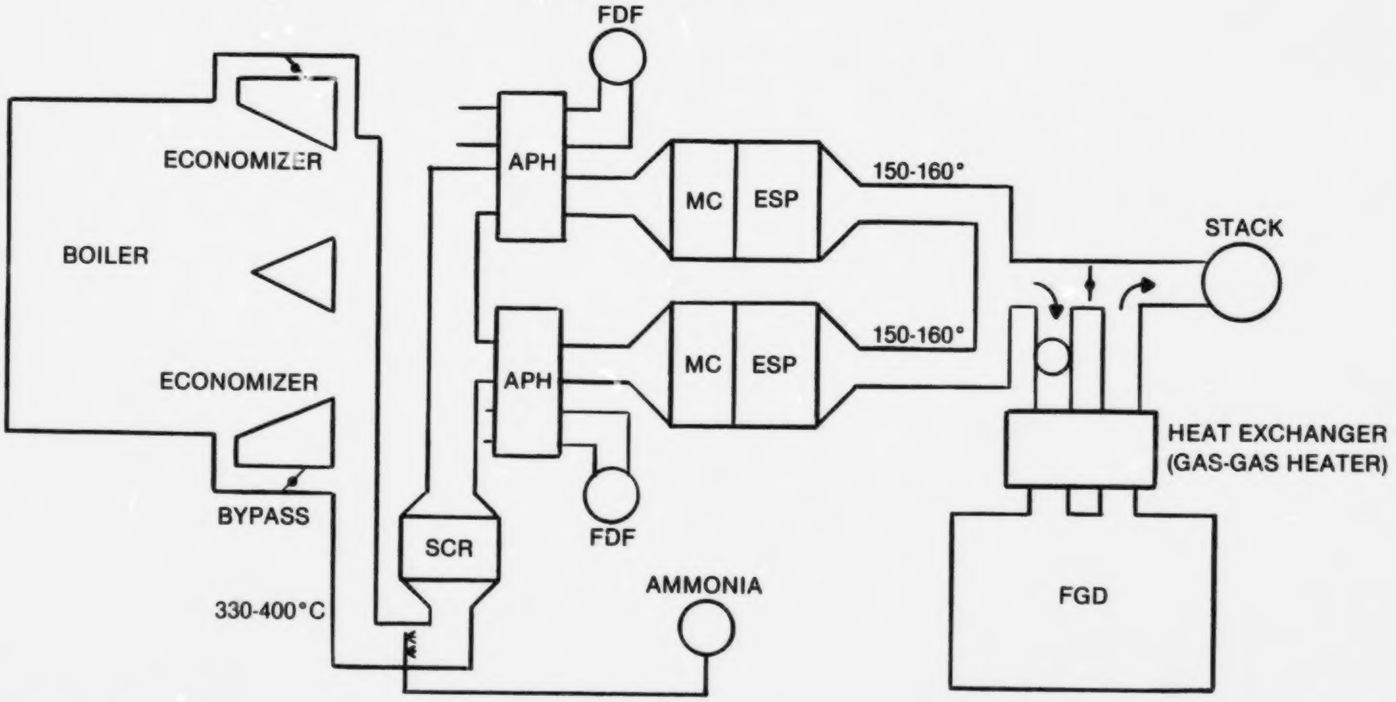
### 4.2.2 SCR System

The SCR reactor at the Shimonoseki Station was retrofitted between the boiler and the air preheater. To meet the regulation, 360°C flue gases containing 500 ppm $NO_x$ are treated with 0.56 mole $NH_3$ per mole $NO_x$ to remove 55% of the $NO_x$. Unreacted ammonia is kept below 1 ppm.

The SCR unit has been operated without any problems for over 2 years since its startup. A catalyst life of over 3 years is expected. The air preheaters downstream of the SCR reactor have had no ammonium bisulfate problem because of the low amount of unreacted $NH_3$ and the high dust concentration.

### 4.2.3 FGD System

A flowsheet of the FGD system is shown in Figure 4-5; Figure 4-6 is a photograph of the unit. The specifications for the scrubbers and other equipment are listed in Tables 4-5 and 4-6.


FDF
ECONOMIZER
BOILER
ECONOMIZER
BYPASS
330-400°C
APH
APH
SCR
FDF
AMMONIA
MC
ESP
MC
ESP
150-160°
150-160°
STACK
HEAT EXCHANGER
(GAS-GAS HEATER)
FGD

SCR: SELECTIVE CATALYTIC REDUCTION OF $NO_x$
MC: MULTICYCLONE
ECO: ECONOMIZER

**Figure 4-4** Flue Gas Treatment System for Shimonoseki Power Station Boiler No. 1, Chugoku Electric.

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## TABLE 4-4. DESCRIPTION OF THE SHIMONOSEKI NO. 1 PLANT

| | |
|---|---|
| **Boiler** | |
| Supplier | MHI |
| Type | Corner firing |
| Capacity, MW | 175 |
| Maximum flue gas, Nm$_3$/hr | 634,000 |
| Fuel | Coal (0.6 - 1.5% S) |
| **Dust Collector** | |
| Type | Multi-cyclone and ESP |
| Supplier | MHI |
| Dust removal efficiency, % | 94-99 |
| **SCR Reactor** | |
| Supplier | MHI |
| Type | Fixed bed |
| Catalyst | Honeycomb |
| $NO_x$ removal efficiency, % | 50 |
| **Gas-Gas Heater (GGH)** | |
| Type | Ljungstrom |
| Supplier | Gadelius Co. |
| **FGD Unit (Scrubber)** | |
| Type | Limestone-gypsum |
| Supplier | MHI |
| $SO_x$ removal efficiency, % | About 96 (after reheating) |
| Dust removal efficiency, % | About 97 (after reheating) |
| Gypsum production, t/hr | 3-8 |
| Wastewater, t/hr | About 9 |

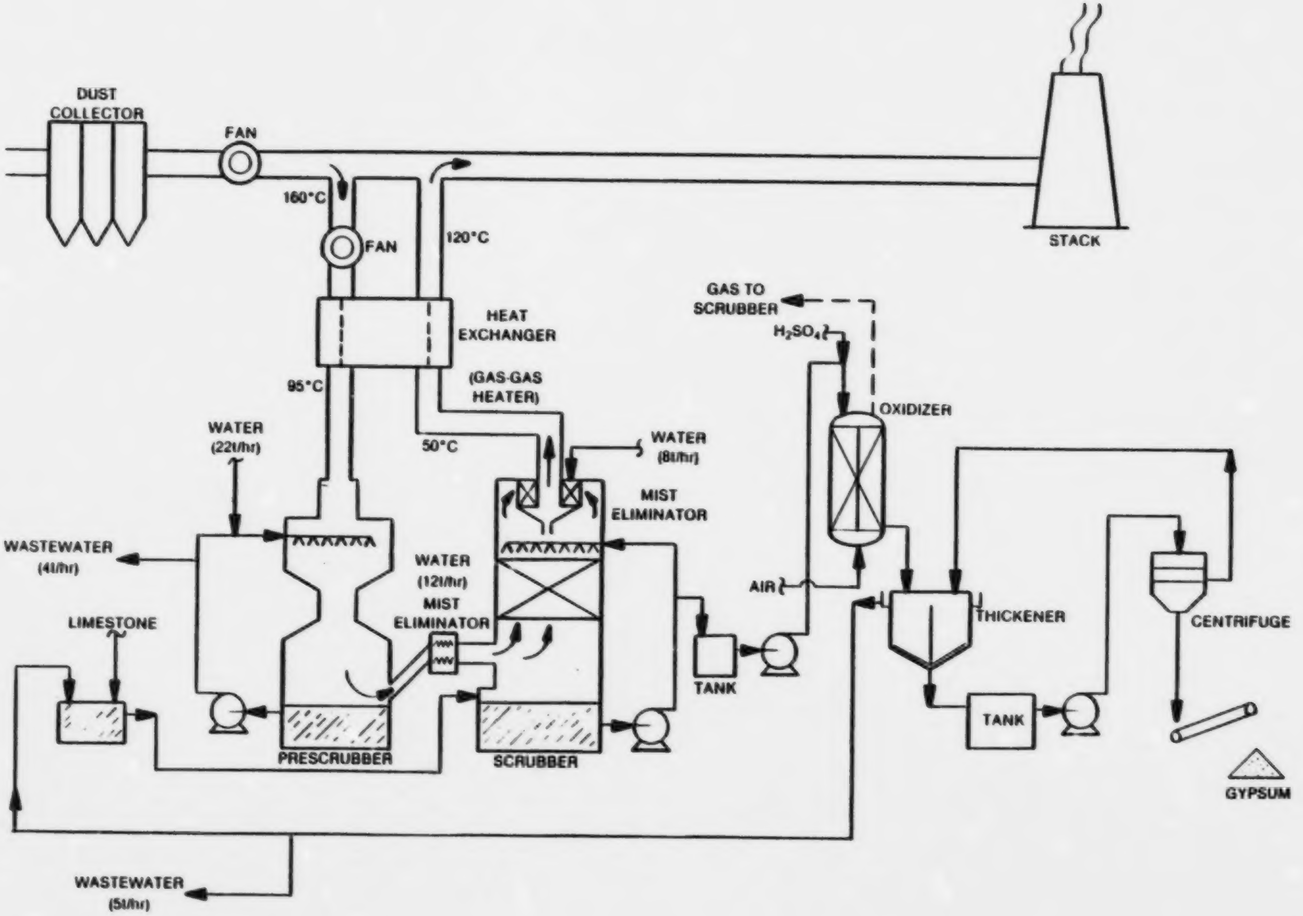
DUST COLLECTOR
FAN
160°C
FAN
120°C
STACK
HEAT EXCHANGER
(GAS-GAS HEATER)
95°C
50°C
GAS TO SCRUBBER
$H_2SO_4$
OXIDIZER
WATER (22t/hr)
WATER (8t/hr)
MIST ELIMINATOR
WASTEWATER (4t/hr)
WATER (12t/hr)
MIST ELIMINATOR
AIR
THICKENER
CENTRIFUGE
LIMESTONE
TANK
TANK
PRESCRUBBER
SCRUBBER
GYPSUM
WASTEWATER (5t/hr)

Figure 4-5 Flowsheet for FGD System on Boiler No. 1, Shimonoseki Power Station.

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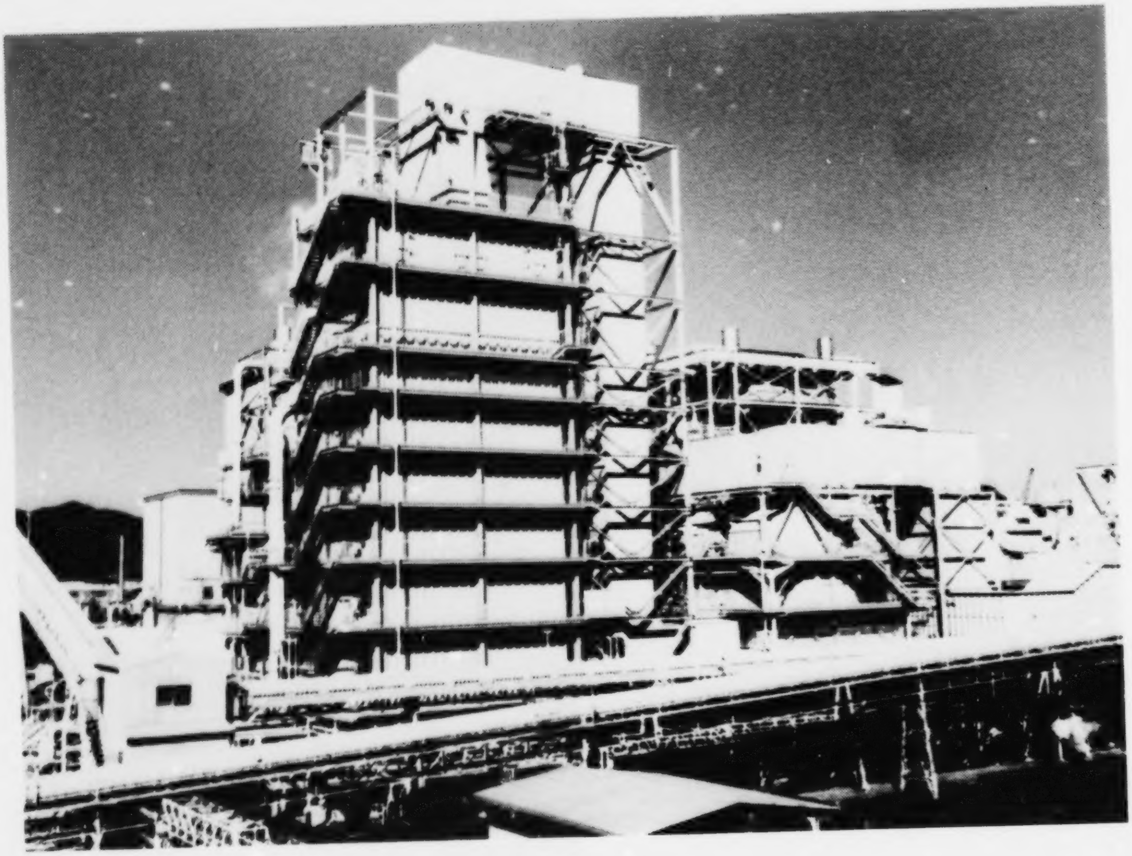

Figure 4-6 FGD Unit at Shimonoseki, Chugoku Electric

## TABLE 4-5. SCRUBBER SPECIFICATIONS

| | |
|---|---|
| Prescrubber (Gas cooling and impurity removal) | |
| Type | Spray tower (semi venturi) |
| Number | 1 |
| Height, m | 30 |
| Shell material | SS 401,410 |
| Lining material | Rubber and resin |
| L/G ratio, l/Nm$^3$ | 3.3 |
| Superficial gas velocity, m/sec | 14 |
| Inlet $SO_2$, ppm | 1500-1700 |
| Inlet particulates, mg/Nm$^3$ | 300-1000 |
| Scrubber | |
| Type | Plastic-grid packed tower |
| Number | 1 |
| Height, m | 38 |
| Shell material | SS 401, 410 |
| Lining material | Resin |
| L/G ratio, l/Nm$^3$ | 14 |
| Superficial gas velocity, m/sec | 2.5 |
| Outlet $SO_2$, ppm | 45-50<br>(100 - 110 at GGH outlet) |
| Outlet particulate, mg/Nm$^3$ | 50-80<br>(60 - 150 at GGH outlet) |
| Pressure drop, mm $H_2O$ | 180 - 240 (total of prescrubber, scrubber, and mist eliminator) |

## TABLE 4-6. SPECIFICATIONS FOR OTHER FGD EQUIPMENT

| | |
|---|---|
| Mist Eliminator | |
| Type | Two-stage Chevron |
| Pressure Drop, mm $H_2O$ | 20-30 |
| Wash Liquor | Fresh water |
| Fan | |
| Capacity, Nm$^3$/m | 700,000 |
| Fan Motor Speed, rpm | 875 |
| Motor Power, kw | 2,490 |
| Thickener | |
| Diameter, m | 10.3 |
| Depth, m | 3 |
| Inlet Solid, % | 13 |
| Outlet Solid, % | 25 |
| Centrifuge | |
| Number | 5 (no spare) |
| Type | Vertical basket |
| Material | SS, rubber lined |
| Capacity, kg | 550 for a batch |
| Inlet Solid, % | 25 |
| Outlet Solid, % | 91 (9 moisture) |

Flue gas leaving the ESP at 150-160°C is cooled to about 95°C by a gas-gas heater and then introduced into a semi-venturi spray tower prescrubber developed by MHI. Most of the particulates, chloride, and fluoride in the gas are removed with the L/G ratio maintained at 3 l/Nm³. About 10 t/hr of wastewater is purged from the prescrubber to remove impurities. Then the gas is passed through a mist eliminator and introduced into the scrubber. The scrubber has plastic-grid packings, a bottom holding tank and a mist eliminator near the exit. Limestone slurry is fed to the scrubber to remove over 90% of the $SO_2$ with an L/G ratio of 14 l/Nm³.

The 50°C treated gas is heated in the gas-gas heater to 110-120°C and sent into the stack. Since about 5% of the untreated flue gas leaks into the treated gas at the heater, both $SO_2$ and particulate removal efficiencies are lowered to around 90% (Table 4-7). Eighty-five percent removal is required to meet the regulations.

TABLE 4-7. $SO_2$ AND PARTICULATE REMOVAL EFFICIENCY (SHIMONOSEKI PLANT)

| Pollutants | Concentration and removal efficiency | |
|---|---|---|
| $SO_2$ | FGD inlet, ppm | 500-820 |
| | FGD outlet, ppm | 20-30 |
| | Removal efficiency, % | 96-96.5 |
| | GGH[a] outlet, ppm | 30-45 |
| | Removal efficiency, % | 94-95 |
| Particulates | FGD inlet, mg/Nm³ | 870-1400 |
| | FGD outlet, mg/Nm³ | 40-50 |
| | Removal efficiency, % | 97-98 |
| | GGH[a] outlet, mg/Nm³ | 70-140 |
| | Removal efficiency, % | 87-95 |

[a] Gas-gas heater.

Leaving the scrubber, the slurry has a pH of about 5. It is treated with sulfuric acid to lower the pH to 4 in the oxidizer where calcium sulfite is air-oxidized to gypsum. The 13% concentration gypsum slurry is thickened to a 25% concentration and then centrifuged to yield byproduct gypsum containing less than about 9% moisture. This gypsum is usually sold for cement and wallboard materials.

The mist eliminator is washed with 8 t/hr of fresh water. Most of the thickener overflow is used to prepare the limestone slurry, a portion is charged to the scrubber and the rest, 5 t/hr, is sent to the wastewater treatment system and treated with water from the prescrubber according to the standard method described in Section 2.4.3.

### 4.2.4 Performance and Economics

After its startup in July 1979, the FGD unit was operated continuously without problems until February 1980, when the boiler was shut down for annual maintenance. During the operation period, coal and oil initially were used in a 25/75 ratio and later in the ratio of 50/50. Inspection during the shutdown period revealed a slight deposit of particulates in the gas-gas heater and a slight erosion of rubber in the pumps, but neither scaling nor corrosion. The soot blowing system was reinforced during the shutdown period to reduce deposit formation in the heater.

The boiler began burning only coal in April 1980, and the FGD unit was operated without problems through July of that year. In August, the FGD unit was shut down for the first time for an 84-hour period to remove fly ash deposits in the heater by water washing. The FGD unit was shut down again in September for about 80 hours because of a large amount of fly ash deposits in the heater caused by an ESP malfunction. The gas-gas heater requires water washing twice a year; washing and drying takes two days. During the wash period the FGD system is shut down and the boiler burns low-sulfur oil. Since its startup in 1979, the FGD system has not been shut down because of scrubber-related problems. Table 4-8 summarizes the performance of the FGD unit.

TABLE 4-8. PERFORMANCE OF FGD UNIT AT SHIMONOSEKI STATION, CHUGOKU ELECTRIC

| Month | Hours |  |  |  | Utilization (%) |  | Oper-ability (C/B,%) | Reli-ability (C/D,%) |
|---|---|---|---|---|---|---|---|---|
|  | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) |  |  |
| June 1980 | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| Aug. | 744 | 744 | 660[a] | 744 | 100 | 88.7 | 88.7 | 88.7 |
| Sept. | 720 | 720 | 641[b] | 720 | 100 | 89.0 | 89.0 | 89.0 |
| Oct. | 744 | 672[c] | 655 | 655 | 90.3 | 88.0 | 100 | 100 |
| Nov. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |
| Dec. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| Jan. 1981 | 744 | 744 | 675[d] | 675 | 100 | 90.7 | 90.7 | 100 |
| Feb. | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 |
| Mar. | 744 | 405[c] | 404 | 404 | 54.4 | 54.3 | 99.8 | 100 |
| Apr. | 720 | 293[c] | 289 | 289 | 40.7 | 40.1 | 98.6 | 100 |
| May | 744 | 740 | 739 | 739 | 99.5 | 99.3 | 100 | 100 |
| June | 720 | 720 | 719 | 719 | 100 | 99.9 | 99.9 | 100 |
| July | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| Aug. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 |
| Sept. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 |

[a] Plugging of gas-heater occurred for the first time since the start of its use in April 1980. The heater was water washed.

[b] ESP malfunction, a large amount of dust deposited in the gas-gas heater requiring water washing.

[c] Boiler was shut down for inspection of the SCR unit.

[d] Inspection of the FGD fan.

The investment cost of the FGD system was 6 billion yen (35,000 yen/Kw) including the gas-gas heater. The cost of the SCR system was 2 billion yen including the boiler modifications needed for installation of the economizer by-pass. The FGD unit has been operated by 3 persons per shift plus 1 additional person during the day shift, while the SCR system requires no operator. The FGD unit consumes about 2.2% of the power generated by the boiler; the SCR system requires about 0.1%.

### 4.2.5 Evaluation

The Shimonoseki plant is the first plant in the world to apply full-scale SCR to a coal-fired boiler. The SCR system has been operated successfully without an adverse effect on the air preheater and FGD units. This plant is also the first to apply full-scale gas-gas heaters to a coal-fired boiler flue gas. The FGD system also has been operated without problems except for dust deposition in the gas-gas heater. The amount of deposits is larger at Shimonoseki than that at the Matsushima plant (Section 3.5.2), presumably because of the larger dust content of the inlet gas--- 200-1300 mg/Nm$^3$ at Shimonoseki versus 50-100 at Matsushima.

The gas-gas heater is fairly costly, but it eliminates the fuel needed for gas reheating and therefore presents a considerable overall economic advantage. $SO_2$ and particulate removal efficiencies are lowered slightly by the gas-gas heater due to gas leakage. Placing the fan between the heater and the FGD unit eliminates the leakage problem, but may cause corrosion of the fan. (Section 2.5). Further improvements in the reheating system are needed to minimize deposition and the lowering of removal efficiencies.

The Shimonoseki plant ably demonstrates that the combination of SCR, ESP, and FGD can make coal flue gas as clean as that from low-sulfur oil.

Because the Shimonoseki plant has proved that the power generation costs of coal-firing with SCR, FGD and ESP are less than those associated with low-sulfur oil or high-sulfur oil and FGD, Chugoku Electric has begun to convert several boilers from oil to coal fuel as shown in Table 1-7. Three units at the Shin-Ube Station recently began operation, using similar SCR-FGD systems to remove 65% of $NO_x$ and 90% of $SO_2$. Unreacted $NH_3$ at the SCR reactors has been maintained at 2 ppm without any adverse effect on the air preheater, FGD system, fly ash, etc. Tests of combined SCR/FGD systems have shown that unreacted $NH_3$ of 5 ppm or less has no adverse effect on the system. Although the Shimonoseki and Shin-Ube units remove 50-65% of $NO_x$, as required by current regulations, tests with these units have demonstrated that 80% $NO_x$ removal can be attained with less than 4 ppm unreacted $NH_3$. If necessary, more catalyst may be added to ensure 80% removal with a lower level of unreacted $NH_3$.

## 4.3 TOMATO-ATSUMA STATION, HOKKAIDO ELECTRIC

### 4.3.1 History

Hokkaido Electric's Tomato-Atsuma Station is located in a large new industrial region in Hokkaido in northern Japan. This station has a new 350 Mw boiler which burns domestic low-sulfur coal (0.3% S) with extensive combustion modification for $NO_x$ abatement including low-$NO_x$ burners, staged combustion and flue gas recirculation. A description of the boiler and air pollution control facilities at the Tomato-Atsuma Station is given in Table 4-9.

The flue gas from the boiler contains about 220 ppm $NO_x$, 220-230 ppm $SO_x$, and nearly 20 g/Nm³ of dust. As shown in Figure 4-7, the gas is first passed through a hot-side ESP to reduce dust to about 25 mg/Nm³ (about 99.9% efficiency). Then one-fourth of the 365°C gas leaving the ESP is subjected to SCR to remove over 80% of $NO_x$ in this stream. The treated gas is then returned to the main flue gas stream. $NO_x$ concentrations in the flue gas are thus maintained below 170 ppm to meet the regulation for $NO_x$.

TABLE 4-9. DESCRIPTION OF THE TOMATO-ATSUMA STATION

| | |
|---|---|
| Boiler capacity, MW | 350 |
| Flue gas volume, $Nm^3/hr$ | 1,268,000 |
| Regulations | |
| Particulate emission limitation, kg/hr | 39 (34 mg/Nm3) |
| $SO_2$ emission limitation, $Nm^3/hr$ | 152 (130 ppm) |
| $NO_x$ emission limitation, $Nm^3/hr$ | 184 (158 ppm) |
| Boiler | |
| Type | Opposed firing |
| Supplier | Babcock-Hitachi Ltd. |
| Service load | Base load |
| Commercial service date | October 31, 1980 |
| Sulfur in coal, % | 0.3 |
| Electrostatic precipitator | |
| Type | Hot side |
| Supplier | Hitachi Plant Engineering and Construction Company |
| Number | 1 (4 compartments) |
| Flue gas temperature, °C | 364 |
| Particulate removal efficiency, % | 99.8 |
| Particulate outlet load, $mg/Nm^3$ | 45 |
| Pressure drop, $mmH_2O$ | 20 |
| FGD | |
| Gas treated, $Nm^3/hr$ | 610,000 |
| Process | Limestone-gypsum |
| Supplier | Babcock-Hitachi Ltd. |
| $SO_2$ removal efficiency, % | Over 90 |
| SCR | |
| Gas treated, $Nm^3/hr$ | 280,000 |
| Supplier | Babcock-Hitachi Ltd. |
| Catalyst | Plate type by Hitachi Ltd. |
| $NO_x$ removal efficiency, % | Over 80 |

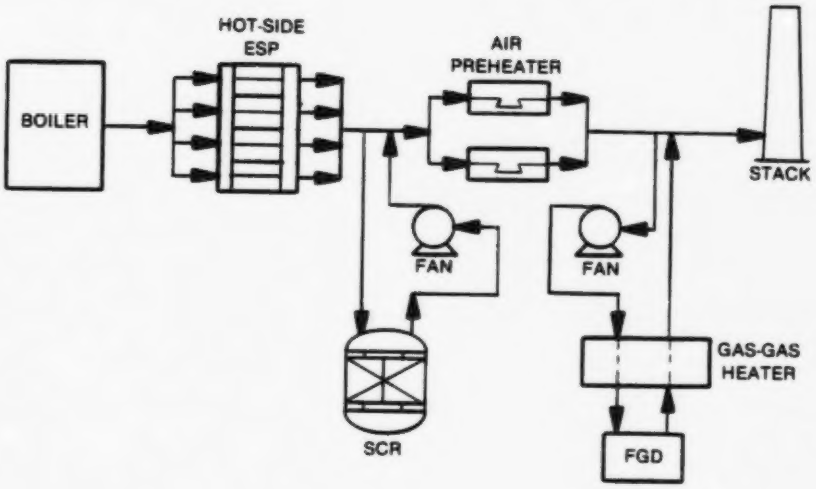
HOT-SIDE ESP
AIR PREHEATER
BOILER
STACK
FAN
FAN
GAS-GAS HEATER
SCR
FGD

Figure 4-7 Boiler and Air Pollution Control Facilities at Tomato-Atsuma Station.

After the gas is cooled to about 150°C in the air preheater, half of it is subjected to a limestone-gypsum process FGD unit to remove over 90% of the $SO_2$; this enables the boiler to meet the regulation for $SO_x$.

The boiler, SCR reactor and FGD unit were constructed by Babcock Hitachi Ltd. and began commercial operation in October 1980. The SCR reactor contains a plate catalyst developed by Hitachi Ltd. The total investment cost of the power station was about 70 billion yen including 11-13 billion yen for air pollution control facilities.

### 4.3.2 FGD Unit

Design conditions and primary equipment of the Tomato-Atsuma FGD unit are shown in Tables 4-10 and 4-11, respectively. A flow sheet of the unit is shown in Figure 4-8. Figure 4-9 is a photograph of the unit. Table 4-12 contains operation data.

In the FGD unit, flue gas with a flow rate of about 610,000 $Nm^3$/hr and a temperature around 140°C is introduced into a Ljungstrom gas-gas heat exchanger. There it is cooled to about 80°C. Once cooled it goes into a venturi prescrubber. The prescrubber uses water to reduce dust to below 10 mg/$Nm^3$; most of the chloride and fluoride in the gas is removed as the water further cools the gas to 50°C. The L/G ratio in the prescrubber is about 2 l/$Nm^3$. The venturi prescrubber produces about 2 t/hr of waste water (pH 1) which is sent to the wastewater treatment system.

Next, the gas is sent through a chevron mist eliminator into a perforated plate scrubber with a low pressure drop. There it is treated with limestone slurry to reduce the $SO_2$ concentration to about 10 ppm. The L/G ratio in the scrubber is 10 l/$Nm^3$ with two pumps, or 5 l/$Nm^3$ with one pump. For low inlet $SO_2$ concentrations of around 230 ppm, one pump is used. The limestone has a purity of about 93% and is finely ground so that 92% of it passes through a 325 mesh screen.

TABLE 4-10. DESIGN CONDITIONS, TOMATO-ATSUMA FGD UNIT

| Items | Design Value | Remarks |
|---|---|---|
| 1. Type | Wet Limestone Gypsum | |
| 2. Gas Flow, Nm$^3$/hr | 610,000 | 350 MW x 1/2 |
| 3. Inlet $SO_2$ ntration, Concentration, ppmv | Maximum 350 | |
| 4. Outlet $SO_2$ Concentration, ppmv | <35 | |
| 5. $SO_2$ Removal, % | >90 (guaranteed by Babcock Hitachi) | at the absorber outlet |
| 6. Inlet Dust Load, mg/Nm$^3$ | <45 | |
| 7. Outlet Dust Load, mg/Nm$^3$ | 15 (guaranteed by Babcock Hitachi) | at the gas-gas heater outlet |
| 8. Gas Reheating | Gas-Gas Heater | |
| 9. Inlet Gas Temperature, °C | 137 | at booster fan inlet |
| 10. Outlet Gas Temperature, °C | 110 | at gas-gas heater outlet |
| 11. Absorbent | Limestone Powder | 325 mesh minus 92% |
| 12. By-products | Gypsum | |
| 13. Operation | Sequential Start and Stop | |
| 14. Fuel | Coal | |

TABLE 4-11. PRIMARY EQUIPMENT SPECIFICATIONS - TOMATO-ATSUMA FGD UNIT

| Equipment | Item | Description |
|---|---|---|
| Booster Fan | Type | Double Suction Turbo Fan |
| | Number | 1 |
| | Flow, $m^3$/min | 17,480 |
| | Pressure, mm $H_2O$ | 760 |
| | Motor, kW | 2,800 |
| Gas-Gas Heater | Type | Vertical Ljungstrom |
| | Number | 1 |
| | Outlet Temperature, °C | 110 |
| Venturi Scrubber | Type | Venturi |
| | Number | 1 |
| | Material | Carbon Steel with Rubber Lining |
| Sump Mist Elminiator | Type | Chevron |
| | Number | 1 |
| | Material | F.R.P. |
| Venti Recirc. Pump | Type | Centrifugal |
| | Number | 2 (1-Stand-by) |
| | Capacity, $m^3$/min/unit | 22.4 |
| | Motor, kW | 200 |
| Absorber | Type | Low Pressure Perforated Plate Tower |
| | Number | 1 |
| | Material | Carbon Steel with Resin Lining |
| Demister | Type | Multi-Pipe |
| | Number | 1 |
| | Material | Carbon Steel with Resin Lining |
| Mist Eliminator | Type | Chevron |
| | Number | 1 |
| | Material | F.R.P. |
| Absorber Recirc. Pump | Type | Centrifugal |
| | Number | 3 (1 standby) |
| | Capacity, $m^3$/min/unit | 51.1 |
| | Motor, kW | 400 |
| Limestone Powder Silo | Type | Vertical Cylindrical |
| | Number | 1 |
| | Capacity, $m^3$ | 230 |

(Continued)

TABLE 4-11. (Continued)

| Equipment | Item | Description |
|---|---|---|
| Oxidizing Tower | Type | Vertical Cylindrical |
| | Number | 1 |
| | Material | Carbon Steel with Resin Lining |
| Air Compressor | Type | Reciprocating |
| | Number | 2 (1-stand-by) |
| | Capacity, $m^3$/min/unit | 12 |
| Thickener | Type | Type with Rake |
| | Number | 1 |
| | Material | Carbon Steel with Rubber Lining |
| Centrifuge | Type | Full-auto, Vertical Basket Type |
| | Number | 3 (1-stand-by) |
| | Capacity, kg/h/unit | 1,240 (dry cake base) |
| | Motor, kW | 30/15 |
| Sodium Carbonate Silo | Type | Vertical Cylindrical |
| | Number | 1 |
| | Capacity, $m^3$ | 24 |
| Settler | Type | Type with Rake |
| | Number | 1 |
| | Material | Carbon Steel with Rubber Lining |

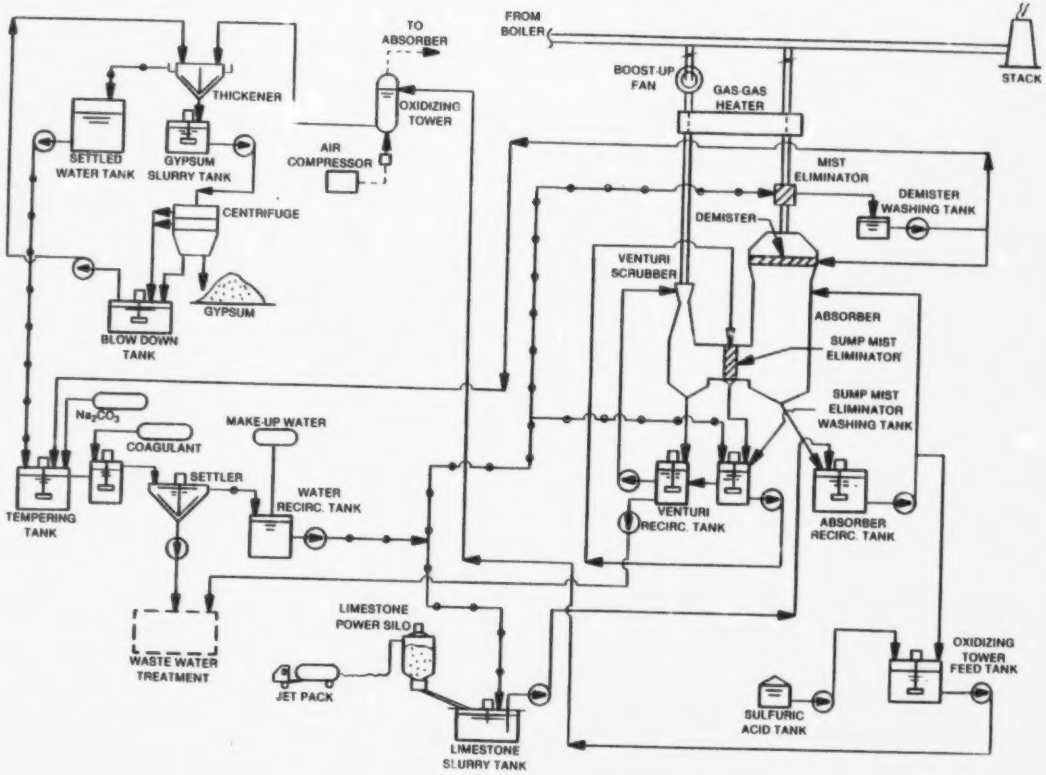
FROM BOILER
STACK
BOOST-UP FAN
GAS-GAS HEATER
MIST ELIMINATOR
DEMISTER WASHING TANK
DEMISTER
VENTURI SCRUBBER
ABSORBER
SUMP MIST ELIMINATOR
SUMP MIST ELIMINATOR WASHING TANK
VENTURI RECIRC. TANK
ABSORBER RECIRC. TANK
OXIDIZING TOWER FEED TANK
SULFURIC ACID TANK
LIMESTONE POWER SILO
JET PACK
LIMESTONE SLURRY TANK
TO ABSORBER
OXIDIZING TOWER
AIR COMPRESSOR
THICKENER
SETTLED WATER TANK
GYPSUM SLURRY TANK
CENTRIFUGE
GYPSUM
BLOW DOWN TANK
$Na_2CO_3$
COAGULANT
MAKE-UP WATER
SETTLER
WATER RECIRC. TANK
TEMPERING TANK
WASTE WATER TREATMENT

**Figure 4-8** Flowsheet of FGD Unit, Tomato-Atsuma Station.

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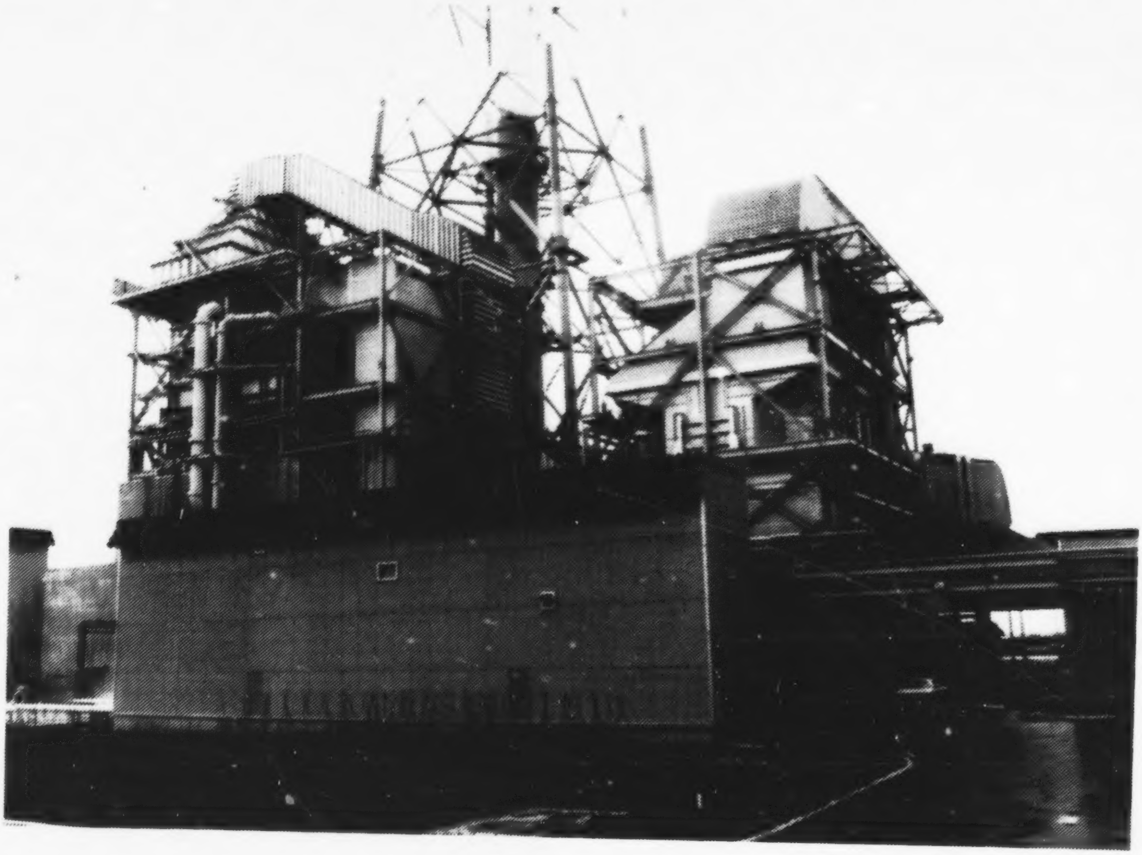


Figure 4-9 FGD Unit at Tomato-Atsuma, Hokkaido Electric

TABLE 4-12. DESIGN DATA AND ACTUAL RESULTS

| Items | Design | Actual | Remarks |
|---|---|---|---|
| Gas flow, $Nm^3/hr$ | 610,000 | 618,000 | at the venturi inlet |
| Inlet $SO_2$ Concentration, ppmv | 350 | 222 | |
| Outlet $SO_2$ Concentration, ppmv | <35 | 8 (at the Absorber Outlet) | 16 ppm at G/G Heater Outlet |
| $SO_2$ Removal Efficiency, % | <90 | 96.4 (at the Absorber Outlet) | 92.8% at G/G Heater Outlet |
| Inlet Dust Load, $mg/Nm^3$ | <45 | 24.4 | |
| Outlet Dust Load, $mg/Nm^3$ | <15 | <5 | |
| Gas Temperature at gas-gas heater, °C | | | |
| (1) Untreated Gas Inlet | 146 | 141.5 | |
| (2) Untreated Gas Outlet | 83 | 81.1 | |
| (3) Treated Gas Inlet | 49 | 50.0 | |
| (4) Treated Gas Outlet | 112 (corrected) | 113.7 | |
| Untreated Gas Leakage, % | 6.3 (initial) | 4.3 | Inlet Gas Basis |
| System Draft Loss, mm $H_2O$ | 525 | 450 | Fan Outlet ~ G/G Heater Outlet |

Note: Since the actual inlet $SO_2$ concentration is lower than that in the design, actual data are based on one absorber recirculation pump operation.

The 50°C gas is then passed through the gas-gas heater where it is heated to 110°C. Since about 4% of the untreated flue gas leaks into the treated flue gas in the heater, the heater outlet gas contains about 16 ppm $SO_2$, lowering the overall $SO_2$ removal efficiency to about 93%. The pressure drop through the gas-gas heater and FGD unit is 450 mm $H_2O$.

The calcium sulfite slurry leaving the scrubber is treated with a small amount of sulfuric acid (about 20 kg/hr) to lower the pH to about 4 and then fed to an oxidizing tower where the sulfite is oxidized to gypsum. The gypsum slurry is treated with a thickener and a centrifuge to yield byproduct gypsum containing about 7% moisture. This is sold for cement and wallboard. The quality and composition of the gypsum are shown in Table 4-13.

TABLE 4-13. BY-PRODUCT GYPSUM

| Items | Analysis Value | Requirement for Cement (Reference) |
|---|---|---|
| Property | | |
| Color | Grayish White | |
| Surface Moisture, % | 6.94 | |
| pH | 6.3 | |
| Composition | | |
| Crystallization Water, % | 20.52 | |
| $CaSO_4 \cdot 2H_2O$, % | 97.6 | Purity: >90% |
| $CaSO_3 \cdot 1/2H_2O$, % | <0.01 | |
| $CaCO_3$, % | 0.06 | <1 |
| $SiO_2$, % | 1.01 | |
| $Fe_2O_3$, % | 0.15 | <2 |
| $Al_2O_3$, % | 0.45 | |
| Cl, % | 0.015 | |

Utility requirements for the FGD unit are shown in Table 4-14. Electric power consumption is 2,880 Kw using one slurry recirculation pump. The consumption is equivalent to 1.65% of 175 MW (half of 350 MW).

TABLE 4-14. UTILITY CONSUMPTION

| Items | Design | Actual | Remarks |
|---|---|---|---|
| Electric Power, HWH/hr | 3,650 | 2,880 | One Absorber Recirc. Pump Operation |
| Industrial Water, t/hr | 14.6 | 10.1 | |
| Limestone (as 100% purity), kg/hr | 900 | 550 | |
| Sulfuric Acid (as 100% purity), kg/hr | 115 | 20 | |
| Sodium Carbonate (as 100% purity), kg/hr | 100 | 40 | |
| Coagulant, kg/hr | 0.12 | 0.06 | |

One of the characteristic features of the Tomato-Atsuma FGD unit is its water softening facility. The gypsum-saturated supernatant of the gypsum slurry thickener is treated with 40 kg/hr of sodium carbonate to remove calcium by precipitation as calcium carbonate. The calcium carbonate is removed as a sludge by a settler. The softened water is then combined with fresh water and used to cool gases in the prescrubber for the preparation of the limestone slurry, and for the mist eliminator wash. By using a softened water feed to the prescrubber, fresh water requirements and wastewater loads from the scrubber system are substantially reduced. The scrubber system purges only 0.21 t/hr of wastewater from the settler.

The amount, quality, and composition of wastewater from the prescrubber and settler are shown in Table 4-15. Those waters are combined, treated with lime to precipitate heavy metals and fluoride, filtered, and then treated by ion exchange to remove dithionate which causes COD.

### 4.3.3 Performance

The boiler has been operated at full load in the daytime and at reduced loads at night with an average capacity factor of 84-85%. As shown in Table

TABLE 4-15. WASTEWATER DESCRIPTION

| Items | Design | Actual |
|---|---|---|
| **Blow-down Water from Venturi Recirc. Tank** | | |
| Blow-down Water, kg/hr | 2,000 | 2,000 |
| pH | 1 ~ 2 | 1.2 |
| SS, ppm | Approx. 7,500 | 7,600 |
| $Cl^-$, ppm | <22,000 | 3,690 |
| $F^-$, ppm | <2,400 | 620 |
| $Na^+$, ppm | Approx. 20,700 | 2,700 |
| $NH_4^+$, ppm | Approx. 500 | 49 |
| COD, ppm | <100 | 77 |
| **Blow-down Water from Settler** | | |
| Blow-down Water, kg/hr | 290 | 210 |
| pH | 9 ~ 9.5 | 9.3 |
| SS, wt% | Approx. 18 | Approx. 0.5 |
| $Cl^-$, ppm | <500 | 460 |
| $F^-$, ppm | <50 | 20 |
| $Na^+$, ppm | <4,900 | 1,360 |
| COD, ppm | Approx. 20 | 20 |

4-16, the FGD unit has been operated with 100% reliability since its startup in October 1980, removing 92-95% of the $SO_2$ at the gas-gas heater outlet. The only problem encountered so far has been scale formation in a pump and in the wastewater treatment system piping. The scale formation has been eliminated by installing a water-wash system for the pump and piping.

### 4.3.4 Evaluation

The Tomato-Atsuma power station is the first one in Japan to use a hot-side ESP system. The ESP has operated well, removing about 99.9% of inlet particulates. The SCR system for $NO_x$ removal has also operated without trouble, removing over 80% of $NO_x$.

The FGD unit has operated with 100% reliability since its startup. Although a minor problem was encountered with the wastewater treatment system, the problem was solved without interrupting scrubber operation.

Babcock Hitachi Ltd. previously constructed an FGD unit for a coal-fired boiler for EPDC at Takehara. The unit underwent a start-up problem causing the shutdown of the FGD unit. The shutdown resulted in a 97.4% reliability rating for the first year. The 100% reliability obtained at the Tomato-Atsuma unit is mainly due to technology improvements and, to a lesser extent, to low inlet $SO_2$ concentrations. The trouble-free operation of the gas-gas heater may be due to the low inlet dust concentration.

Due to the use of a perforated plate scrubber with a larger hole area, the pressure drop through the Tomato-Atsuma FGD system is much lower than that of Takahara (Table 2-3). The energy requirement at Tomato-Atsuma is low (1.7% of the power generated by the boiler). This low energy consumption is a result of good design as well as low inlet $SO_2$ concentrations.

Water softening requires a few simple facilities and sodium carbonate as a feedstock but reduces the fresh water requirements as well as the wastewater load. It may also support reliable scrubber operations.

### TABLE 4-16. PERFORMANCE OF FGD UNIT AT TOMATO-ATSUMA STATION, HOKKAIDO ELECTRIC

| Month | Hours | | | | Utilization (%) | | Oper-ability (C/D,%) | Reli-ability (C/D,%) | $SO_2$ removal (%) |
|---|---|---|---|---|---|---|---|---|---|
| | Total (A) | Boiler operation (B) | FGD operation (C) | FGD called to operate (D) | Boiler (B/A) | FGD (C/A) | | | |
| Oct., 80[a] | 744 | 523 | 523 | 523 | 70.3 a | 70.3 a | 100 | 100 | 94.4 |
| Nov. | 720 | 613 | 613 | 613 | 85.1 | 85.1 | 100 | 100 | 92.4 |
| Dec.[b] | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 94.1 |
| Jan., 81 | 744 | 712 | 712 | 712 | 95.7 | 95.7 | 100 | 100 | 95.2 |
| Feb. | 672 | 672 | 672 | 672 | 100 | 100 | 100 | 100 | 93.3 |
| Mar. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.1 |
| Apr. | 720 | 719 | 719 | 719 | 99.9 | 99.9 | 100 | 100 | 93.3 |
| May | 744 | 639 | 736 | 636 | 85.9 | 85.5 | 99.5 | 100 | 95.3 |
| June | 720 | 701 | 699 | 699 | 97.4 | 97.1 | 99.7 | 100 | 95.6 |
| July | 744 | 744 | 737 | 737 | 99.1 | 99.1 | 100 | 100 | 94.2 |
| Aug. | 744 | 744 | 744 | 744 | 100 | 100 | 100 | 100 | 93.7 |
| Sept. | 720 | 720 | 720 | 720 | 100 | 100 | 100 | 100 | 93.5 |

[a] Boiler began commercial operation in October 1980.

[b] Minor trouble occurred with wastewater treatment facility but was solved without shutdown of scrubbers.

Although the sodium content of the scrubber liquor exceeds 1,000 ppm, sodium seems to produce no adverse effects on the scrubbing operation or on the gypsum quality.

The Tomato-Atsuma unit's byproduct gypsum has a purity of over 97%. This purity is due to the highly efficient hot-side ESP and to the prescrubber which removes most impurities in the gas. The gypsum grows into large thick crystals, contains only 7% moisture after being centrifuged, and should be suitable for use in cement and wall-board.

The SCR reactor upstream of the FGD units adds about 5 ppm of $NH_3$ due to leakage. Since about one-fourth of the total flue gas is subjected to SCR, the flue gas at the FGD inlet may contain about 1 ppm $NH_3$, which is caught by the prescrubber. The ammonia content of the wastewater from the prescrubber is low--only about 50 ppm--and so far no treatment for ammonia removal is required.

As mentioned above, the total air pollution control system at Tomato-Atsuma has been working very well with a high removal efficiency. However, the investment cost is fairly high. The cost for the total air pollution control system for the 350 MW boiler was 11-13 billion yen and accounts for 15.7-18.6% of the total investment cost of the power station. The cost is high considering that the FGD unit treats half of the flue gas while the SCR unit treats only one-fourth of the gas.

The high investment cost may be due to the conservative but complex design used to achieve high removal efficiencies and high reliabilities. The low $SO_2$ emission from the scrubber (8 ppm) may be the lowest figure yet achieved for wet limestone processes for coal-fired boiler operations. Operation of the unit with 100% reliability since startup is an excellent record. The relatively small sizes of the FGD unit (175 MW equivalent) and SCR unit (88 MW equivalent) may also be one of the reasons for the relatively high cost per kW. Although the operating cost seems fairly low, further reduction of the investment cost is desirable.

# SECTION 5

# OTHER FGD PROCESSES FOR COAL

## 5.1 INTRODUCTION

While limestone-gypsum processes have been applied predominantly to coal-fired boilers, several modified lime/limestone processes also have been developed for coal-fired boiler applications (Table 5-1). The major features of these processes are shown in Table 5-2.

The standard limestone-gypsum process as described in the previous sections requires good operational control to prevent scaling. Although automatic control systems have been used on new units, well-trained operators are still needed. Processes that give no trouble even with relatively rough control are desirable, particularly for smaller units. The modified lime/limestone processes reduce the possibility of scaling by using additives (Nos. 1, 3, 4, and 5 in Table 5-1) or by lowering the pH (No. 2). Other targets of the processes are low investment cost, low power consumption, and a small amount of wastewater. The processes require no sulfuric acid except Process No. 5 in Table 5-2. Processes 3, 4, and 5 may not require a prescrubber to remove chloride derived from the coal.

Three processes (No. 1, 4, and 5 of Table 5-2) use lime with additives and attain a high $SO_2$ removal efficiency with low power consumption (Processes 1 and 4) or eliminate wastewater (Process 5). The lime cost, however, may increase with the fuel cost.

Process No. 6 uses red mud, a useless byproduct of aluminum production from bauxite. With this process, red mud is utilized for $SO_2$ removal before it is discarded. Although the red mud process may be less costly its application is probably limited to FGD units near aluminum plants.

TABLE 5-1. OTHER FGD PROCESSES FOR COAL

| No. | Absorbent | By-Product | Process Developer | Commercial Application | Status of Application for Coal |
|---|---|---|---|---|---|
| 1 | CaO, MgO | Gypsum | KHI | Oil-fired boiler | Commercial in 1981 in West Germany, in 1983 in Japan |
| 2 | $CaCO_3$ | Gypsum | Chiyoda | Oil-fired boiler | 23 MW test in U.S. |
| 3 | $Al_2(SO_4)_3$, $CaCO_3$ | Gypsum | Dowa | Various sources | 10 MW test in U.S. |
| 4 | CaO, $CH_3COONa$ | Gypsum | Kureha | Oil-fired boiler | 1.5 MW test in Japan |
| 5 | CaO, $CaCl_2$ | Gypsum | Kobe Steel | Sintering machine | 1.5 MW test in Germany |
| 6 | Red Mud | Sludge | Sumitmo Aluminum | Oil-fired boiler | Possible commercial |

## TABLE 5-2. CHARACTERISTICS OF FGD PROCESSES FOR COAL

(●: Excellent; o: Good; #: Average; Δ: Poor)

| | 1 KHI | 2 Chiyoda | 3 Dowa | 4 Kureha | 5 Kobe | 6 Sumitomo |
|---|---|---|---|---|---|---|
| Ease of operation control | o | o | o | o | # | # |
| $SO_2$ removal efficiency | o | # | # | ● | o | o |
| Capital cost | o | o | o | # | # | ● |
| Power consumption | o | o | o | o | # | o |
| Reduction of wastewater | o | # | # | # | ● | # |
| Saving of reheating energy | # | # | # | # | o | # |
| Economy of absorbent | Δ | o | # | Δ | Δ | ● |
| Quality of by-product | o | o | o | o | o | Δ |

All of these processes are described in this section.

In addition, dry FGD processes using activated carbon have been tested by the Federation of Electric Power Companies on flue gas from coal in an attempt to produce sulfur as a byproduct (Table 5-3). To date there is no available information on these studies. An activated carbon process using ammonia for simultaneous $SO_x$ and $NO_x$ removal and the electron beam process will be described in Chapter 6. Semi-dry lime/limestone processes using spray dryer reactors will not be used in Japan because of the difficulty involved in disposing of the byproduct which consists of calcium sulfite, sulfate, lime/limestone, and fly ash.

TABLE 5-3. TEST UNITS OF CARBON PROCESS FGD FOR COAL BY FEDERATION OF ELECTRIC POWER COMPANIES

| Plant Site | Power Company | Constructor | Gas treated ($Nm^3$/hr) |
|---|---|---|---|
| Omura | Kyushu Electric | Mitsubishi Heavy Industries | 30,000 |
| Ebetsu | Hokkaido Electric | Babcock Hitachi K. K. | 10,000 |
| Naie | Hokkaido Electric | Ishikawajima-Harima Heavy Industries | 10,000 |

## 5.2 MAGNESIUM-GYPSUM FGD PROCESS

### 5.2.1 Development Status

Kawasaki Heavy Industries (KHI) developed the magnesium-gypsum process and has constucted two units for oil-fired boilers. Recently KHI constructed a unit to treat one-third of the flue gas from a 700 MW coal-fired boiler at the Mehrum Station, D.B.A., West Germany; it began operation in September 1981.

KHI also is constructing magnesium-gypsum FGD units for two coal-fired boilers (156 MW and 250 MW) at Shikoku Electric's Saijo Station. They will be ready for operation in 1983 and 1984, respectively.

### 5.2.2 Process Description

The magnesium gypsum process for coal-fired boilers is similar to that used at the Japan Exlan plant which treats 300,000 $Nm^3/hr$ of flue gas from a 100 MW oil-fired boiler(11). The flowsheet of the Japan Exlan unit is shown in Figure 5-1. $SO_2$ is absorbed by a slurry containing $Mg(OH)_2$, $Ca(OH)_2$, and gypsum. $Mg(OH)_2$ and $Ca(OH)_2$ react with $SO_2$ to form $MgSO_3$, $CaSO_3$, $Mg(HSO_3)_2$, and $Ca(HSO_3)_2$. Air is introduced into the slurry to oxidize the sulfites and bisulfites to sulfates and bisulfates. The slurry is treated by a thickener and centrifuge to separate the gypsum byproduct. The mother liquor is treated with lime to produce a slurry containing $Mg(OH)_2$, $Ca(OH)_2$, and gypsum which is sent to the absorber. The relationship of the pH and L/G ratio to the $SO_2$ removal efficiency is shown in Figure 5-2. Magnesium increases the $SO_2$ removal efficiency and prevents gypsum scaling.

The Japan Exlan unit has no prescrubber. Although the mist eliminator is washed with fresh water to prevent gypsum scaling, no wastewater is normally purged because water is evaporated in the scrubber.

The specifications for the units used with coal-fired boilers are shown in Table 5-4 (12). The Mehrum unit is designed to treat one-half of the gas from the boiler (1,224,000 $Nm^3/hr$) and has been treating one-third of the gas (816,000 $Nm^3/hr$). The unit has no prescrubber and purges 20t/hr of wastewater from the scrubber system to maintain a chloride concentration of 10,000 ppm in the scrubber liquor. The byproduct gypsum contains about 100 ppm chloride after being water washed in the centrifuge, and is used for wall board.

The Saijo units have a prescrubber with a separate liquor loop in order to obtain higher quality gypsum and a higher $SO_2$ removal efficiency. The flue gases at Saijo contain less chloride but much more fluoride than does

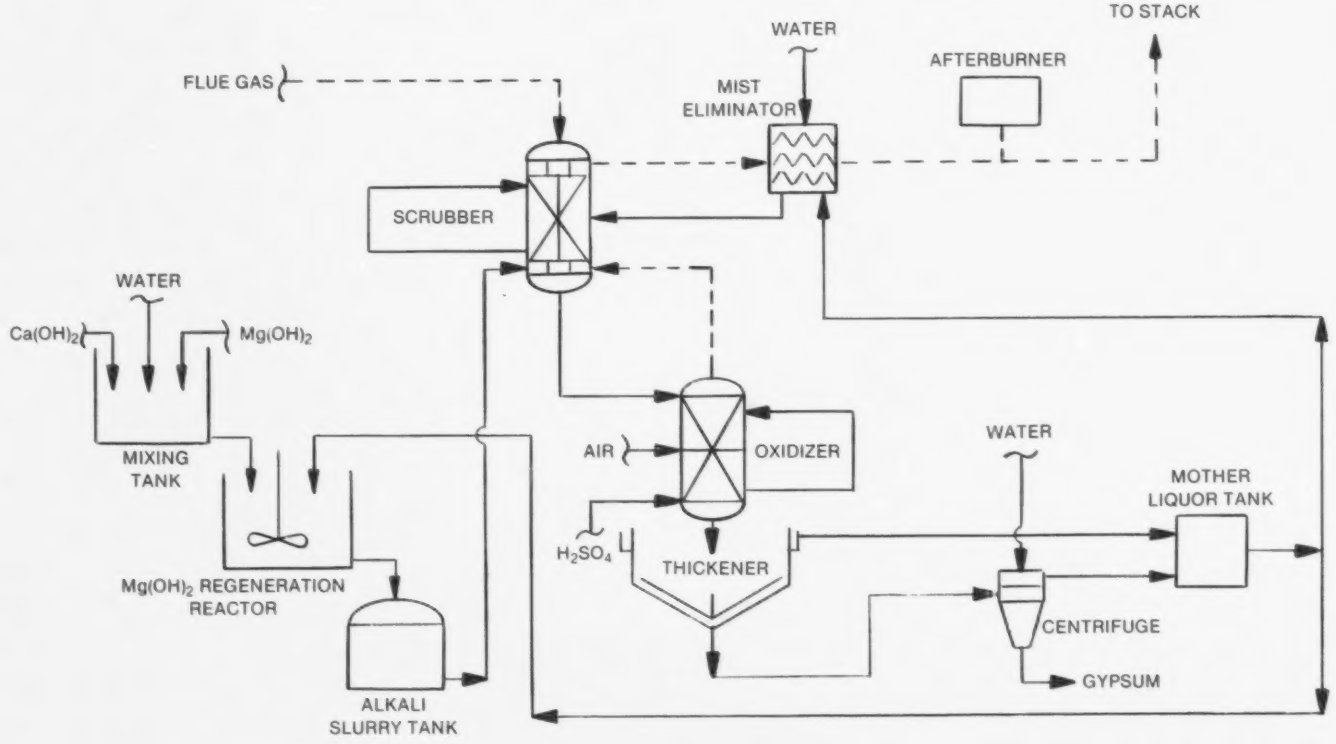
 
FLUE GAS
WATER
MIST ELIMINATOR
AFTERBURNER
TO STACK
SCRUBBER
WATER
$Ca(OH)_2$
$Mg(OH)_2$
MIXING TANK
$Mg(OH)_2$ REGENERATION REACTOR
ALKALI SLURRY TANK
AIR
OXIDIZER
$H_2SO_4$
THICKENER
WATER
MOTHER LIQUOR TANK
CENTRIFUGE
GYPSUM

Figure 5-1 Flowsheet of the Magnesium-Gypsum FGD Process Used with the Japan Exlan Oil-Fired Boiler.

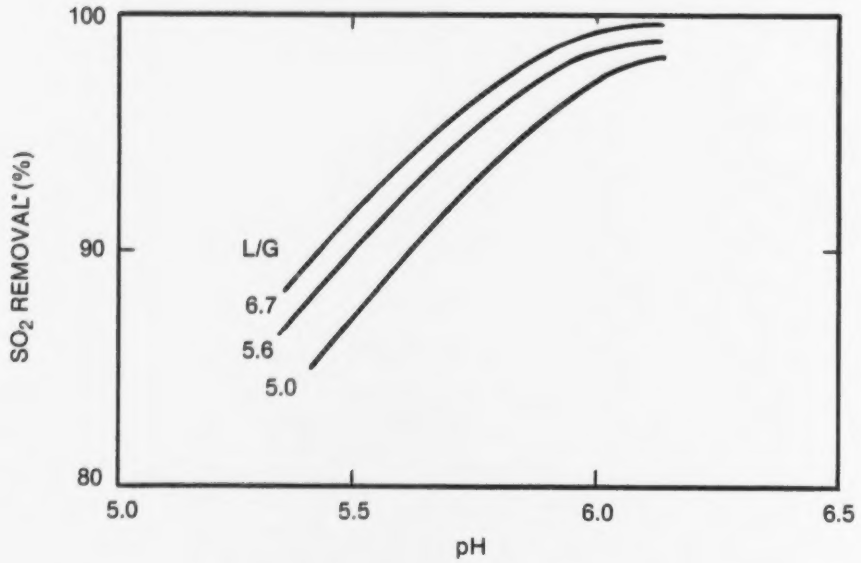


**Figure 5-2** **Absorbent pH and $SO_2$ Removal. (KHI Process)**

TABLE 5-4. SPECIFICATIONS OF FGD UNITS FOR COAL (MAGNESIUM-GYPSUM PROCESS) (12)

| Item | Mehrum No. 3 | Saijo No. 1 | Saijo No. 2 |
|---|---|---|---|
| Startup Date | Sept. 1981 | April 1983 | Oct. 1983 |
| Unit Output, MW | 700 | 156 | 250 |
| Treating Ratio | 1/3 | 1/1 | 1/1 |
| Treating Gas Volume, $Nm^3/h$ | 816,000 $Nm^3/hr$ | 557,000 $Nm^3/hr$ | 858,000 $Nm^3/hr$ |
| Inlet Concentration (design value) | | | |
| $SO_2$, ppm | 851 (wet) | 820 (dry) | 914 (dry) |
| HCl, ppm | 95 (wet) | 34 (dry | 36 (dry) |
| HF, ppm | 12 (wet) | 74 (dry) | 79 (dry) |
| Particulate, $mg/Nm^3$ | 100 (wet) | 100 (dry) | 100 (dry) |
| Outlet Concentration (design value) | | | |
| $SO_2$, ppm | 110 (Absorber) | 82 (stack) | 91 (stack) |
| Particulate, $mg/Nm^3$ | <100 (Absorber) | 30 (stack) | 30 (stack) |
| $SO_2$ removal efficiency, % | 87 (Absorber) | 90 (overall) | 90 (overall) |
| Particulate efficiency, % | — | 70 (overall) | 70 (overall) |
| Alkali | CaO | $Ca(OH)_2$ | $Ca(OH)_2$ |
| Magnesium | $MgSO_4 \cdot H_2O$ | $Mg(OH)_2$ slurry | $Mg(OH)_2$ slurry |
| By-product | Gypsum | Gypsum | Gypsum |
| $CaSO_4 \cdot 2H_2O$ (dry) | 4.6 ton/hr | 3.1 ton/hr | 5.0 ton/hr |
| Effluent Liquor, $m^3/h$ | 20 | 12 | 7 |
| Sulfuric Acid | None | None | None |
| Reheating Method | After burning | Gas-gas heater (2 trains) | Gas-gas heater (2 trains) |
| Reheating Temp., °C | 95 | 98 | 98 |
| Pre-scrubber | None | 1 | 1 |
| Size (dia. x height, m) | - | 9.76 x 21 | 12.14 x 33 |
| Type | - | Spray tower | Spray tower |
| Absorber | 1 | 1 | 1 |
| Size (dia. x height, m) | 14.5 x 32.5 | 9.76 x 31 | 12.14 x 33 |
| Type | Spray tower | Spray tower | Spray tower |
| Mist Eliminator | Chevron type | Chevron type | Chevron type |
| Use for | Absorber | Pre-scrubber<br>Absorber | Pre-scrubber<br>Absorber |
| Recirculation Pump | | | |
| for Pre-scrubber | -<br>- | 3 (1)[a]<br>680$m^3$/hr x 40m | 3 (1)[a]<br>1,025$m^3$/hr x 40m |
| for Absorber | 3 (1)[a]<br>2,900$m^3$/hr x 45m | 4 (1)[a]<br>1,220$m^3$/hr x 45m | 4 (1)[a]<br>1,890$m^3$/hr x 45m |
| Oxidizer | 1 | 2 (series) | 2 (series) |
| Centrifuge | 4 (1)[a] | 4 (1)[a] | 5 (1)[a] |
| Type | Basket | Basket | Basket |

[a] The number in parenthesis indicates standby.

the gas at Mehrum. The units have a water-softening system to reduce wastewater; the purge liquor from the scrubber is treated with sodium hydroxide and carbonate to precipitate magnesium and calcium, and the supernatant is sent into the prescrubber.

### 5.2.3 Economics and Evaluation

The construction cost of the Saijo FGD units was about 20,000-25,000 yen/kW excluding civil engineering costs (12). The cost includes the price of a gas-gas heat exchanger which accounts for 10% of the total amount. The total is slightly less than that for the standard limestone-gypsum process unit. The small size of the slurry circulating pumps and the lack of a pH controller (with sulfuric acid) contribute to the low cost. KHI estimates the cost to be about 6% lower when the prescrubber is not used.

The operating costs of the magnesium-gypsum FGD units for coal are shown in Table 5-5 (12). The process requires a make-up magnesium compound which costs about 5% of what lime costs and is much less expensive than the sodium compound used for water softening.

The magnesium-gypsum process requires lime, which is more expensive than limestone. On the other hand, the process is characterized by scale-free operation and low power consumption (1.6-2.0% of the power generated by the coal-fired boiler with 800 ppm $SO_2$)(11). Although magnesium tends to hinder the crystal growth of gypsum, KHI has developed a method of obtaining a good quality salable gypsum. Those advantages may compensate for the cost disadvantage of the lime requirement.

## 5.3 JET BUBBLING FGD PROCESS

### 5.3.1 Development Status

Chiyoda Chemical Engineering and Construction Co. has developed an FGD process using a jet bubbling scrubber and limestone absorbent (9). The process was demonstrated in the U.S. at Gulf Power's Scholz Plant in Florida

TABLE 5-5. OPERATING COST OF MAGNESIUM GYPSUM PROCESS UNITS (20)

| Item | Mehrum | Saijo No. 2 | | |
|---|---|---|---|---|
| 1. Treating capacity, MW | 700 x 1/3 (233) | 250 x 1/1 | | |
| 2. Gas flow rate, $Nm^3/hr$ | 816,000 | 858,000 | | |
| 3. Treating $SO_2$, kg-mol/hr | 27.0 | 28.4 | | |
| 4. Utility | Consumption | Consumption | Unit Price | Cost |
| $CaO$ | 1,520 kg/hr | - | | |
| $Ca(OH)_2$[a] | - | 2,170 kg/hr | 17 yen/kg | 36,890 yen/hr |
| $MgSO_4 \cdot H_2O$ | 57 t/hr | - | | |
| $Mg(OH)_2$ | - | 26 kg/hr | 70 yen/kg | 1,820 yen/hr |
| Fresh Water | 57 kg/hr | 33 Ton/hr | 30 yen/kg | 990 yen/hr |
| $NaOH$[b] | - | 44 kg/hr | 150 yen/kg | 6,600 yen/hr |
| $Na_2CO_3$[b] | - | 32 kg/hr | 70 yen/kg | 2,240 yen/hr |
| Coagulant | 50 g/hr | 40 g/hr | 2 yen/g | 80 yen/hr |
| Fuel oil | 1,350 kg/hr | - | - | |
| Steam[c] | - | 2,700 kg/hr | 3 yen/kg | 8,100 yen/hr |
| Electricity | 3,750 kw | 4,900 kw | 15 yen/kwhr | 73,500 yen/hr |
| | | | | 130,220 yen/hr |
| 5. By-product ($CaSO_4 \cdot 2H_2O$) | 4,650 kg/hr | 4,970 kg/hr | | |
| 6. Effluent Liquor | 20 $m^3$/hr | 12 $m^3$/hr | | |
| 7. Operating personnel | 3 operators per shift | 3 operators per shift take care of both No. 1 and No. 2 FGD plants. | | |

[a] Quality of $Ca(OH)_2$ is 90 percent passed through 325 mesh.
[b] For a water softening treatment.
[c] For soot blowing of gas-heaters.

over a 10 month period beginning in August 1978. The unit treats 95,000 $Nm^3$/hr of coal flue gas containing 800 - 3,500 ppm $SO_2$. Chiyoda has constructed a commercial unit for an oil-fired boiler belonging to Mitsubi-shi Petrochemical at Yokkaichi which began operation in May 1982. The unit has a capacity of treating 260,000 $Nm^3$/hr of flue gas containing 2,000 ppm $SO_2$ at a 95% removal efficiency. Chiyoda is likely to receive an order for 2 200 MW coal-fired utility boilers in Japan, and will probably use this FGD system with a prescrubber.

### 5.3.2 Process Description (13)

A flowsheet of the prototype jet bubbling process at the Scholz Power Plant is shown in Figure 5-3. Ninety-five thousand $Nm^3$/hr of flue gas from the coal-fired boiler is passed through an ESP for dust removal and then is introduced into a prescrubber where the gas is cooled by water spray. In the jet bubbling reactor, the gas is dispersed into liquid, forming a foam layer. Limestone slurry is fed to the reactor so that flue gas $SO_2$ reacts with limestone and air to form gypsum. The gypsum slurry is withdrawn from the reactor and disposed of in a gypsum stack. The liquid clarified in the stacking pond is returned to the system for reuse.

### 5.3.3 Operation of the Scholz Plant (13)

Table 5-6 summarizes the operating conditions during a 10-month test operation period at the Scholtz station. Bottled $SO_2$ was added when an $SO_2$ level of over 2,000 ppm was required for test purposes. The operation was closely evaluated by the Electric Power Research Institute (EPRI) (14). Results indicated that the reliability of the system was over 99.5% during the 10 months; the average limestone utilization was 98.5%. The $SO_2$ removal efficiency is shown in Figure 5-4.

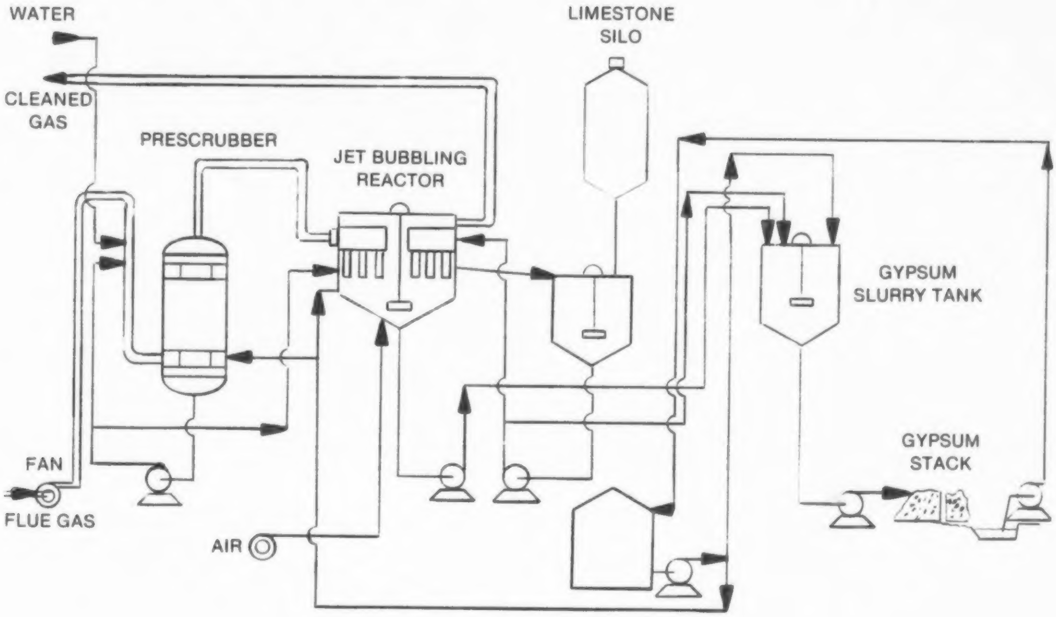

WATER
CLEANED GAS
PRESCRUBBER
JET BUBBLING REACTOR
LIMESTONE SILO
GYPSUM SLURRY TANK
GYPSUM STACK
FAN
FLUE GAS
AIR

**Figure 5-3** Jet Bubbling FGD Process at Gulf Power's Scholz Station.

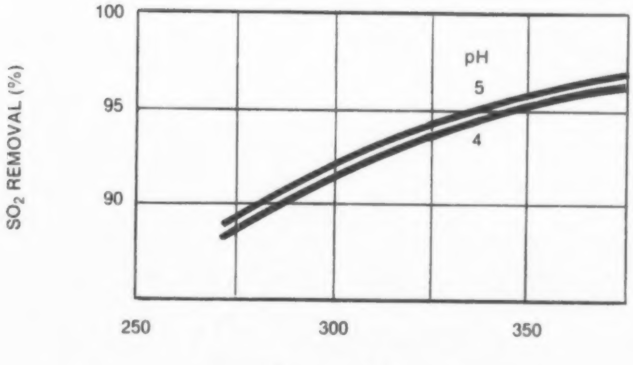


**Figure 5-4** $SO_2$ Removal as a Function of Pressure Drop. (as predicted by equation (Radian Corp.) for inlet $SO_2$ = 2200 ppm).

TABLE 5-6. SUMMARY OF OPERATING CONDITIONS OF CT-121 PROTOTYPE PLANT

| | |
|---|---|
| Flue gas flow, $Nm^3/hr$ | 42,000 - 95,000 |
| Inlet $SO_2$, ppm | 800 - 3,500 |
| Pressure drop across reactor, mm $H_2O$ | 150 - 380 |
| Absorbent pH | 2.5 - 5.0 |
| Oxidation air stoichiometry | 2.0 - 9.0 |
| Limestone size | 90% pass 200 and 325 mesh |

The jet bubbling reactor had a high particulate removal efficiency during the test period. When the ESP was intentionally deactivated so that the flue gas leaving the precipitator contained about 10 $g/Nm^3$ of particulates, over 99.5% of the particulates were removed.

During the 10 month operation period the plant internals were inspected four times but were not cleaned because no significant deposits or scale had formed. The mist eliminator located at the flue gas exit duct was washed with pond water once every two weeks for about one minute to remove soft deposits. No hard scale formed.

Figures 5-5 and 5-6 summarize the results of the laboratory tests. These tests indicated that the $SO_2$ removal efficiency was lowered at higher inlet $SO_2$ concentrations and that magnesium sulfate, 2-5% in the absorbent slurry, significantly increased the $SO_2$ removal efficiency.

### 5.3.4 Yokkaichi Plant, Mitsubishi Petrochemical

The first commercial plant to use the jet bubbling scrubber was completed in April 1982 at Mitsubishi Petrochemical's Yokkaichi works. The plant has a capacity of treating 250,000 $Nm^3/hr$ of flue gas from an industrial boiler burning 3-4% sulfur oil. The flue gas contains 1500-2000 ppm $SO_2$ and 200 $mg/Nm^3$ of dust. Table 5-7 lists the major equipment used in the plant.

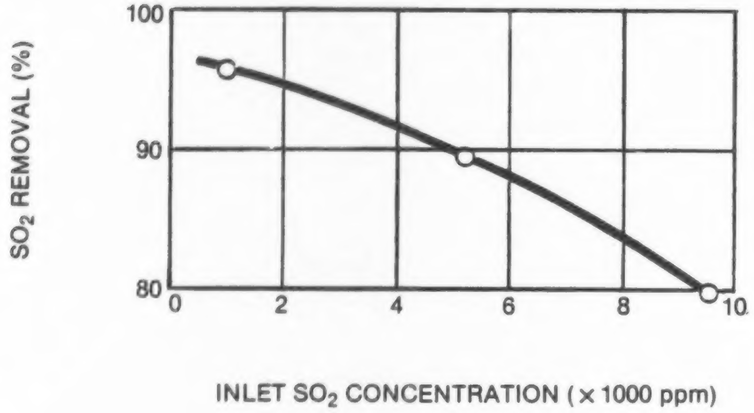


**Figure 5-5** Relationship between $SO_2$ Removal and Inlet $SO_2$ Content. (Jet bubbling reactor pressure drop = 300 mm $H_2O$, 900 Ø reactor, pH = 5).

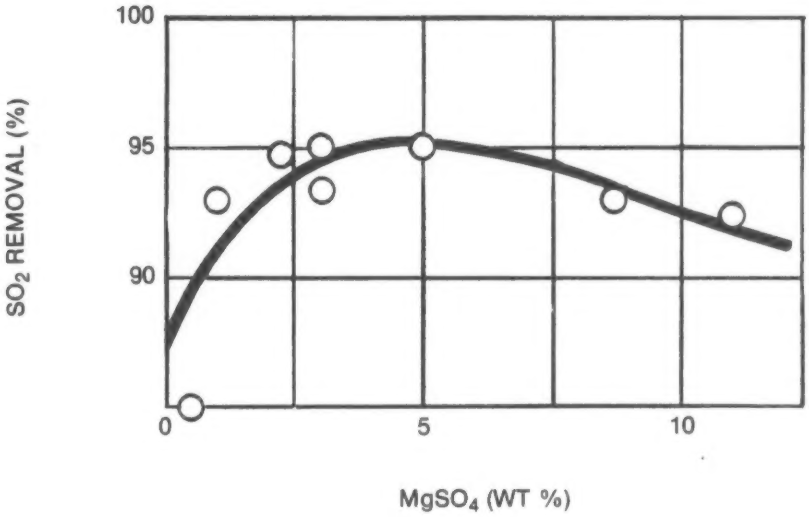


**Figure 5-6** Relationship between $SO_2$ Removal and $MgSO_4$ Content. (Inlet $SO_2$ = 1000 ppm, pH = 6, jet bubbling reactor pressure drop = 300 mm $H_2O$).

## TABLE 5-7. MAJOR EQUIPMENT (YOKKAICHI UNIT)

| Item | Quantity | Specification | Material |
|---|---|---|---|
| Flue gas fan | 1 | Turbo fan<br>256,00 $Nm^3$/hr | Carbon steel |
| Pre-scrubber | 1 | Venturi+Separator | Carbon steel+ FRP lining |
| Jet bubbling reactor | 1 | Cylindrical type<br>9.8mϕ x 9.7mH<br>Agitator<br>Paddle | SUS-316L+ FRP solid |
| Thickener | 1 | Cylindrical<br>4.0mϕ x 5.1mH | Carbon steel +Rubber lining |
| Oxidation Air-Fan | 1+1Spare | Turbo-fan<br>5000 $Nm^2$/hr | Carbon steel |
| Limestone Powder storage | 1 | Vertical-cylindrical type (for 4 days)<br>6.8mϕ x 19.6mH | Carbon steel |
| Centrifuge | 3 | Super decanter<br>Fully automatic<br>1.33t/h | SUS-316L |
| Mist eliminator | 1 | Chevron type<br>2 Stages | |

The plant has operated without problems since startup, removing 97% of $SO_2$ and 85% of dust using limestone ground so that 90% of it passes through a 325 mesh screen. Total pressure drop through the FGD plant is 500 mm $H_2O$ including 400 mm $H_2O$ in the scrubber. The power consumption is about 1200 kW (equivalent to about 1.5% of the total power generated by the boiler). The plant consumes 20 t/hr of water and purges 3 t/hr of wastewater.

The investment cost was 1.5 billion yen (equivalent to 18,000 yen/kW) excluding civil engineering costs. The plant has been operated by two operators per shift and by-produces over 97% pure gypsum which is sold for wallboard production.

### 5.3.5 Evaluation and Economics

The jet bubbling process is useful for producing salable gypsum. To obtain high quality gypsum with flue gas from a coal-fired boiler, the liquor leaving the prescrubber should be purged after being treated by a conventional method (Section 2.3.4). The process is simple. It requires neither slurry recirculation pumps at the scrubber nor sulfuric acid or a separate oxidizer to promote oxidation.

The investment cost of a jet bubbling FGD unit for coal is about 22,000 yen per kW. The cost may be slightly higher than that of the simple throw-away sulfite sludge unit, but is probably less than that of a conventional limestone-gypsum process unit. The power consumption is fairly low--about 1.7% of the power generated by the boiler with coal may be used for the gypsum recovery process. For a throw-away gypsum process, the power consumption may be slightly lower than this.

## 5.4 ALUMINUM SULFATE - LIMESTONE FGD PROCESS

### 5.4.1 Development/Application Status

Dowa Mining Company developed the aluminum sulfate - limestone FGD process to recover gypsum and constructed 11 commercial plants of this type, including one in China (11). The process has been licensed to Universal Oil Products (UOP Inc.) in the U.S. Pilot plant tests of the process with flue gas from coal were carried out from late 1979 through June 1980 at the Tennessee Valley Authority's (TVA) Shawnee Station. By modifying an existing pilot plant, TVA was able to achieve a capacity of treating 25,000 acfm of flue gas at 300°F (about 27,000 $Nm^3$/hr). The scrubber and oxidizers used were different from the standard types and some of the test results were not satisfactory. Tests were conducted again in early 1982 at Shawnee using standard scrubbers and oxidizing towers.

### 5.4.2 1980 Tests at Shawnee (15)

A flowsheet of the Shawnee unit used for testing of the aluminum sulfate-limestone process is shown in Figure 5-7. Flue gas containing about 3,000 ppm $SO_2$ and below 0.1 g/$sft^3$ of particulates at 300°F was introduced into a turbulent contact absorber (TCA) scrubber and treated with the basic aluminum sulfate solution ($Al_2(SO_4)_3 \cdot Al_2O_3$) at a pH of about 3.5. The solution contained 20 g/l of aluminum. The liquor leaving the scrubber was treated in a holding tank by air bubbling to oxidize the aluminum sulfite formed in the scrubber to aluminum sulfate. The liquor was then reacted with limestone in two-stage reactors to precipitate gypsum. The gypsum was removed and the mother liquor containing the basic aluminum sulfate was returned to the scrubber.

The operation was carried out without scaling problems. Scale that had formed in the scrubber system during previous tests was dissolved by the aluminum sulfate liquor. However, the $SO_2$ removal efficiency was lower than desired--87% with the L/G ratio of 52 gal/$kft^3$ (about 7.5 l/$Nm^3$). It was found that the gas distribution was poor in the scrubber and caused the low efficiency.

The scrubber was subsequently modified and fixed-bed polygrid packing was placed at depths of 6 feet and 9 feet. In this way, stable operation with fairly high $SO_2$ removal efficiencies was attained when the gas volume was 20,000 acfm or below. For example, 93% $SO_2$ removal was attained at

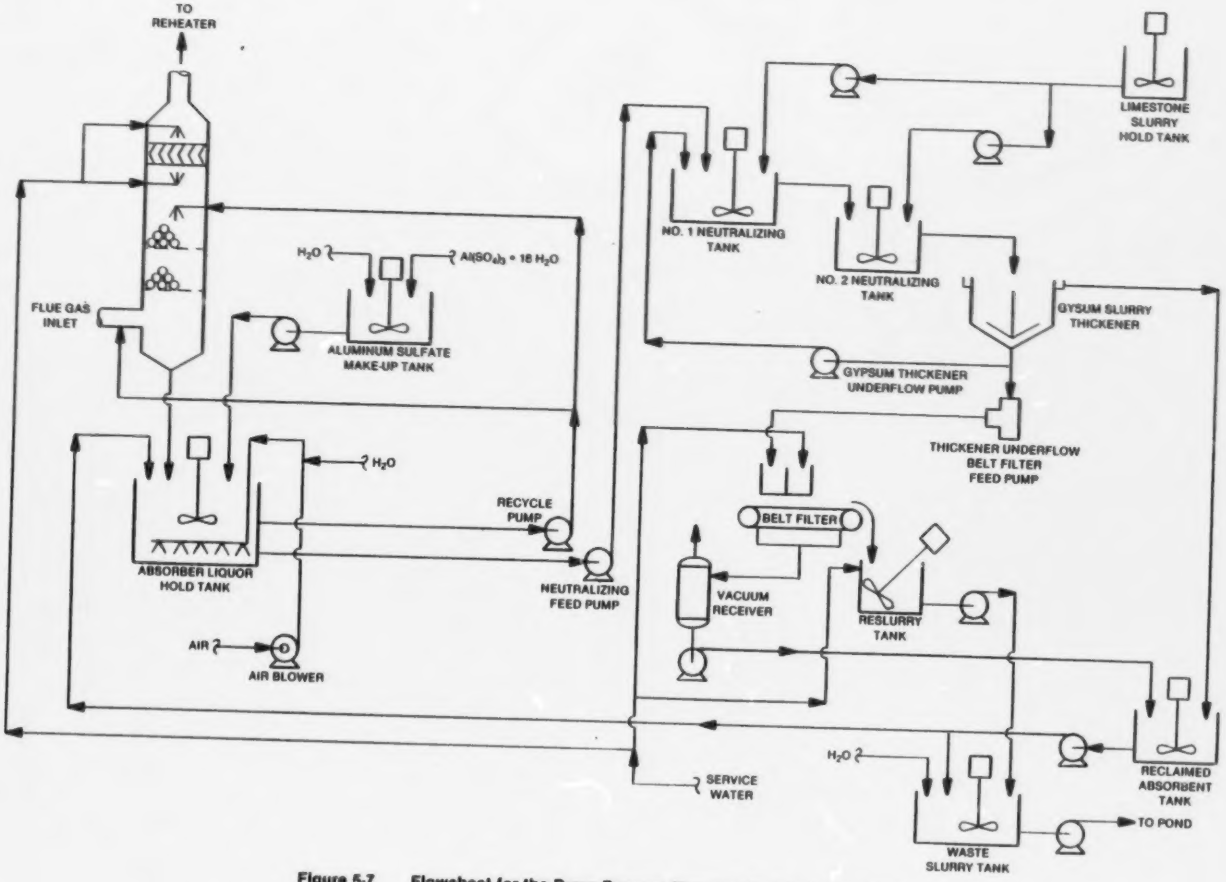

TO REHEATER
FLUE GAS INLET
$H_2O$
$Al_2(SO_4)_3 \cdot 18 H_2O$
ALUMINUM SULFATE MAKE-UP TANK
$H_2O$
ABSORBER LIQUOR HOLD TANK
RECYCLE PUMP
NEUTRALIZING FEED PUMP
AIR
AIR BLOWER
NO. 1 NEUTRALIZING TANK
NO. 2 NEUTRALIZING TANK
LIMESTONE SLURRY HOLD TANK
GYSUM SLURRY THICKENER
GYPSUM THICKENER UNDERFLOW PUMP
THICKENER UNDERFLOW BELT FILTER FEED PUMP
BELT FILTER
VACUUM RECEIVER
RESLURRY TANK
SERVICE WATER
$H_2O$
WASTE SLURRY TANK
RECLAIMED ABSORBENT TANK
TO POND

**Figure 5-7** **Flowsheet for the Dowa Process Shawnee Test Unit. (1980 test).**

20,000 acfm (superficial gas velocity of about 9 ft/min.), an L/G ratio of 82 gal/kft$^3$ (about 12 l/Nm$^3$) and a pressure drop of 9 in $H_2O$ (230 mm), while 93-97% was achieved with a 13,000 acfm flow, an L/G of 90-120, and a pressure drop of 0.5-2.5 in. $H_2O$. The relationship of the gas velocity and rate of absorbing liquor to $SO_2$ removal and pressure drop is shown in Figure 5-8.

The results are still not as good as those expected from experiences with the process in Japan.

### 5.4.3 1982 Tests at Shawnee (16)

The Shawnee plant was modified to be similar to the original Dowa process (Figure 5-9), and operated for several months in early 1982. Test conditions were chosen to reflect typical operation conditions expected in a full-scale commercial Dowa system. The range of variables and design points for the tests is shown in Table 5-8. Aluminium was recovered from wastewater by adding lime and precipitating aluminum hydroxide.

TABLE 5-8. SHAWNEE PLANT TEST CONDITIONS (1982 test)

| | |
|---|---|
| Flue gas flow rate, acfm at 300°F | 16,500-27,500 |
| Absorber superficial gas velocity, fps | 7.5-12.5 |
| Inlet $SO_2$ concentration, ppm | 1,500-2,500 (uncontrolled) |
| Inlet particulate concentration, gr/dscf | 0.4-0.8 |
| Oxidation air rate, scfm | 120-140 |
| Liquor rate to absorber, gpm | 600-830 |
| Absorber packing height, ft. | 10(6' + 4') |
| Nominal aluminum concentration, g/L | 20 |
| Absorber liquor basicity, % | 10 |
| Regenerated liquor basicity, % | 30 |
| Redissolution tank basicity, % | 15 |
| Solids concentration in neutralization tank, wt. % | 10 |
| Thickener underflow solids concentration, wt. % | 35 |
| Feed to aluminum recovery system, gpm | 1.5-2.9 |

Test results using 10 feet of packing are summarized in Table 5-9. The L/G ratio required to achieve 90% $SO_2$ removal was substantially reduced without much increase in the pressure drop. High quality gypsum was produced.

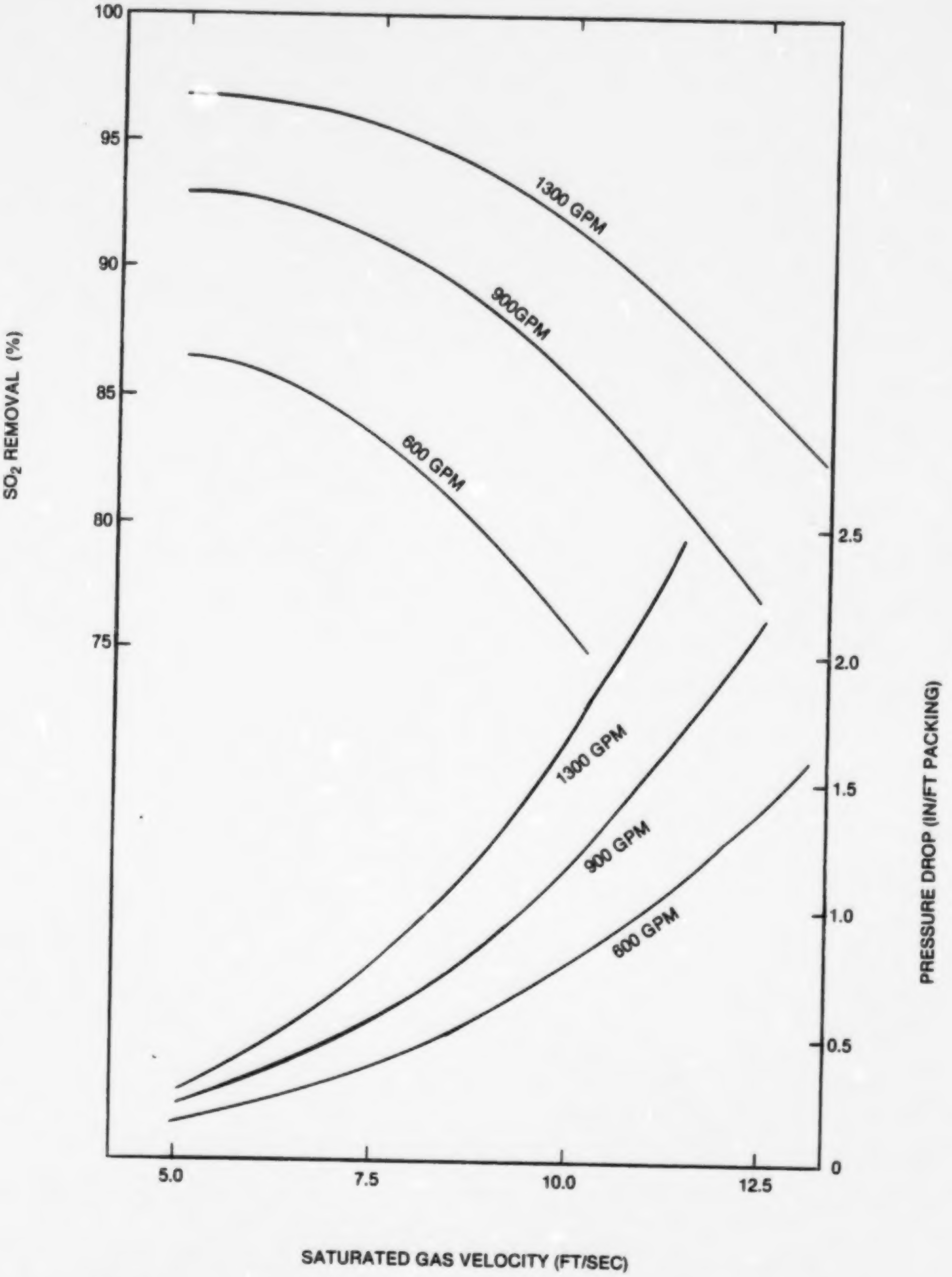


**Figure 5-8** **Characteristics of Polygrid Packed Scrubber. (6 foot packing, 1980 test).**

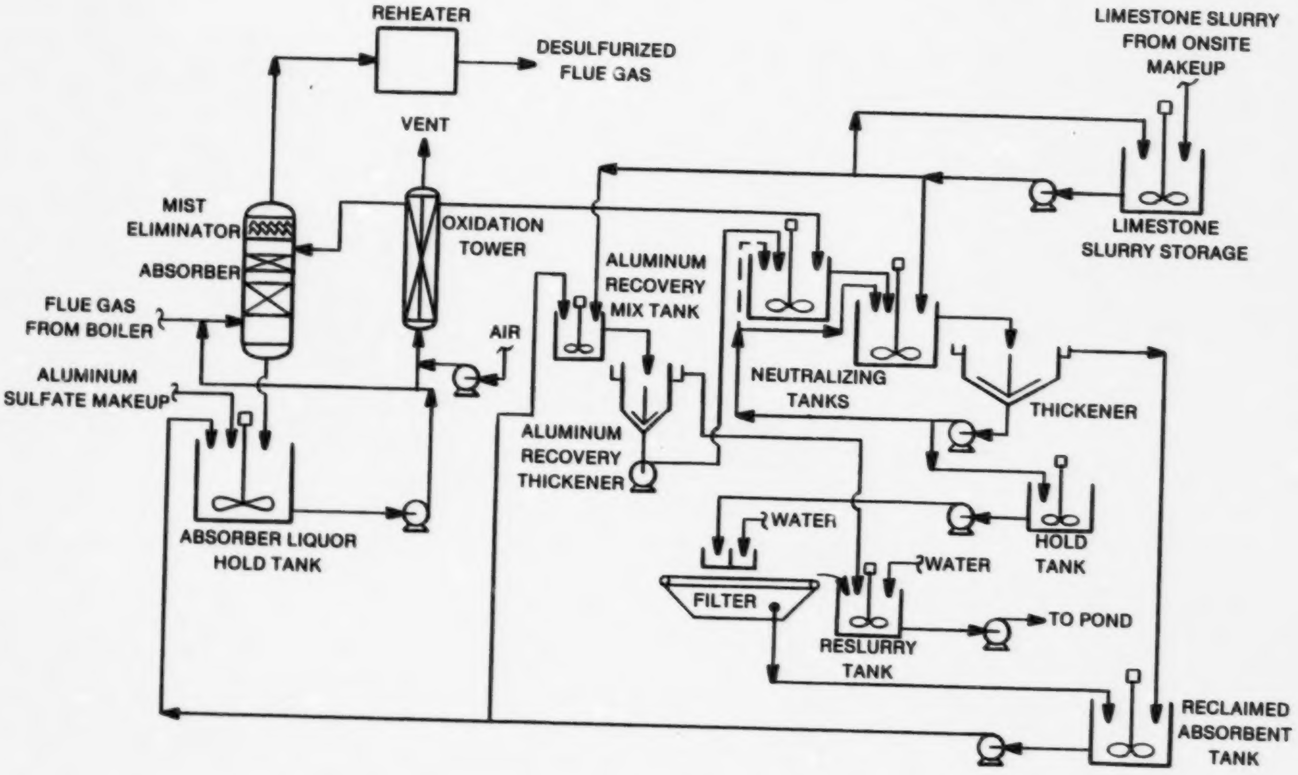

REHEATER
DESULFURIZED FLUE GAS
VENT
LIMESTONE SLURRY FROM ONSITE MAKEUP
MIST ELIMINATOR
ABSORBER
FLUE GAS FROM BOILER
OXIDATION TOWER
AIR
ALUMINUM RECOVERY MIX TANK
NEUTRALIZING TANKS
LIMESTONE SLURRY STORAGE
ALUMINUM SULFATE MAKEUP
ALUMINUM RECOVERY THICKENER
THICKENER
ABSORBER LIQUOR HOLD TANK
WATER
WATER
HOLD TANK
FILTER
RESLURRY TANK
TO POND
RECLAIMED ABSORBENT TANK

Figure 5-9 Dowa Process Flow Diagram - Shawnee Test Facility. (1982 test)

TABLE 5-9. SHAWNEE PLANT TEST RESULTS (1982 test)

| | DSD-7 | DSD-8 | DSD-9 | DSD-10 | DSD-11[a] | DSD-12 |
|---|---|---|---|---|---|---|
| Absorber gas rate, acfm at 250°F | 23,800 | 24,500 | 19,100 | 19,100 | 26,000 | 16,500 |
| Absorber superficial gas velocity, fps | 10.8 | 11.1 | 8.7 | 8.7 | 12.5 | 7.5 |
| Liquor rate, gpm to absorber packing | 640 | 750 | 640 | 690 | 740 | 640 |
| L/G, gas/kcf saturated | 31 | 36 | 39 | 42 | | 45 |
| Limestone Stoichiometry | 1.0 | 1.0 | 1.0 | 1.0 | | 1.0 |
| $SO_2$ removal, % | 83 | 87 | 92 | 94 | | 95 |
| Absorber ΔP, inches w.c. | 5.0 | 8.8 | 2.5 | 2.7 | | 2.0 |

[a] Attempts were made to run at 12.5 fps absorber superficial gas velocity with this packing but flooding conditions were encountered.

TABLE 5-10. AVAILABILITY OF SHAWNEE PLANT (1982 test)

$$\text{Availability} = \frac{\text{Hours Dowa system operated}}{\text{Period hours-Outage hours not caused by Dowa process}}$$

| WEEK | AVAILABILITY % | REASON FOR SYSTEM BEING DOWN |
|---|---|---|
| 2/1 | 100.0 | |
| 2/8 | 100.0 | |
| 2/15 | 100.0 | |
| 2/22 | 100.0 | |
| 3/1 | 100.0 | |
| 3/8 | 81.8 | Repair G-201 recycle pump |
| 3/15 | 100.0 | |
| 3/22 | 93.2 | Repair G-201 recycle pump seal water leak |
| 3/29 | 100.0 | |
| 4/5 | 98.1 | Repair G-201 recycle pump bearing |
| 4/12 | 75.8 | Finish repairs on G-201 pump bearing |
| 4/19 | 100.0 | |
| Overall Availability | 95.7 | |

The availability of the plant is shown in Table 5-10. The plant was operated with a high degree of reliability. Pump bearing and seal problems were the primary cause of downtime. It should be noted that if spare pumps had been installed, as they are in a commercial system, the outages would not have affected plant availability.

### 5.4.4 Evaluation

The Dowa limestone process is characterized by scale-free operation, high quality gypsum and no need for sulfuric acid. The Shawnee test demonstrated the scale-free operation of the process for coal-fired applications.

The L/G ratio, 39 gal/kft$^3$ (6 l/Nm$^3$) needed to attain a 92% $SO_2$ removal efficiency is not large for a single scrubber treating flue gas containing about 1,500-2,500 ppm $SO_2$. For 1,000 ppm $SO_2$, the L/G ratio may be reduced to 30-35 gal/kft$^3$. The ratio may be further reduced when a prescrubber is used to lower the gas temperature or when a two-scrubber system is employed.

With flue gas from coal, chloride accumulates in the scrubber liquor, which necessitates purging of the wastewater. For commercial operation, it may be preferable to recover aluminum from the wastewater. Aluminum sulfate has been widely used for wastewater treatment and produces no adverse effect on the environment.

## 5.5 CALCIUM CHLORIDE-LIME-GYPSUM FGD PROCESS

### 5.5.1 Development/Application Status

Kobe Steel has developed an FGD process which uses a 20% $CaCl_2$ solution containing CaO as the absorbent. The company has constructed several commercial units mainly for flue gas from iron-ore sintering units. Lime is about 7 times more soluble in the chloride solution than in water (Figure 5-10), allowing a smaller L/G ratio - 3 versus 10. Since the vapor pressure of the chloride solution is much lower than that of water (Figure 5-11), the scrubber outlet gas temperature can be higher - 70° versus 55°C, necessitating less gas reheating and cooling water. The high chloride operation is suitable for treating coal flue gas which contains a considerable amount of chloride.

The process has been licensed to Steag of Uhde, West Germany. Steag has conducted pilot plant testing of the process using 5,000 Nm³/hr of flue gas from a coal-fired boiler.

### 5.5.2 Process Description and Commercial Plant Operation

A flowsheet of the process is shown in Figure 5-12. Flue gas at 150°C is contacted in a prescrubber with an approximately 5% $CaCl_2$ solution for cooling and for concentration of the solution to about 20%. The concentrated solution is fed to the scrubber, combined with lime, and used to remove about 90% of $SO_2$. The calcium sulfite slurry is treated in a thickener. The thickened slurry is combined with sulfuric acid and water and then contacted with air bubbles for oxidation of the sulfite to gypsum. The water is added in order to dilute the chloride concentration to about 5%, thereby increasing the oxidation rate. The gypsum is centrifuged and washed with water to yield a gypsum byproduct containing about 200 ppm chloride. This gypsum can be used for cement and wallboard.

Operation data for a commercial unit at Kobe Steel's Kakogawa Works are shown in Table 5-11. The Kakogawa unit has been operated since May 1978. Initially the unit had scaling problems with the mist eliminator when circulating liquor was used for the wash. The scaling problem has since been eliminated by using fresh water to wash the eliminator. Fresh water wash requirements account for less than 1/40 of the amount of circulating liquor previously used for the wash. Virtually no wastewater is purged. A small amount of make-up $CaCl_2$ is needed because the flue gas contains little chloride.

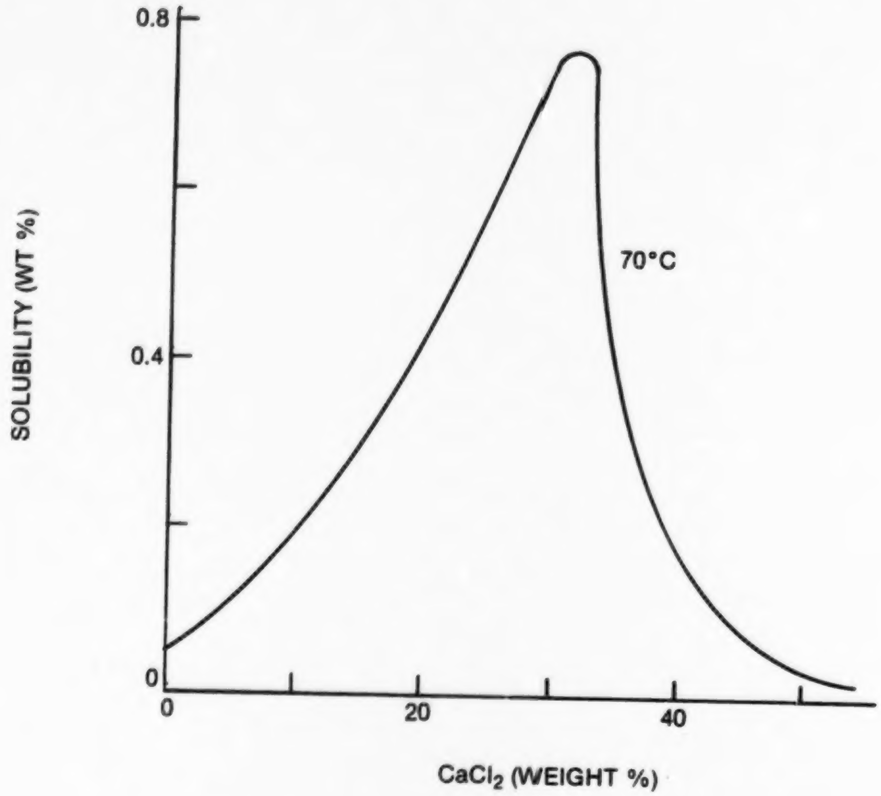


**Figure 5-10** **Solubility of $Ca(OH)_2$ in $CaCl_2$ Solution.**

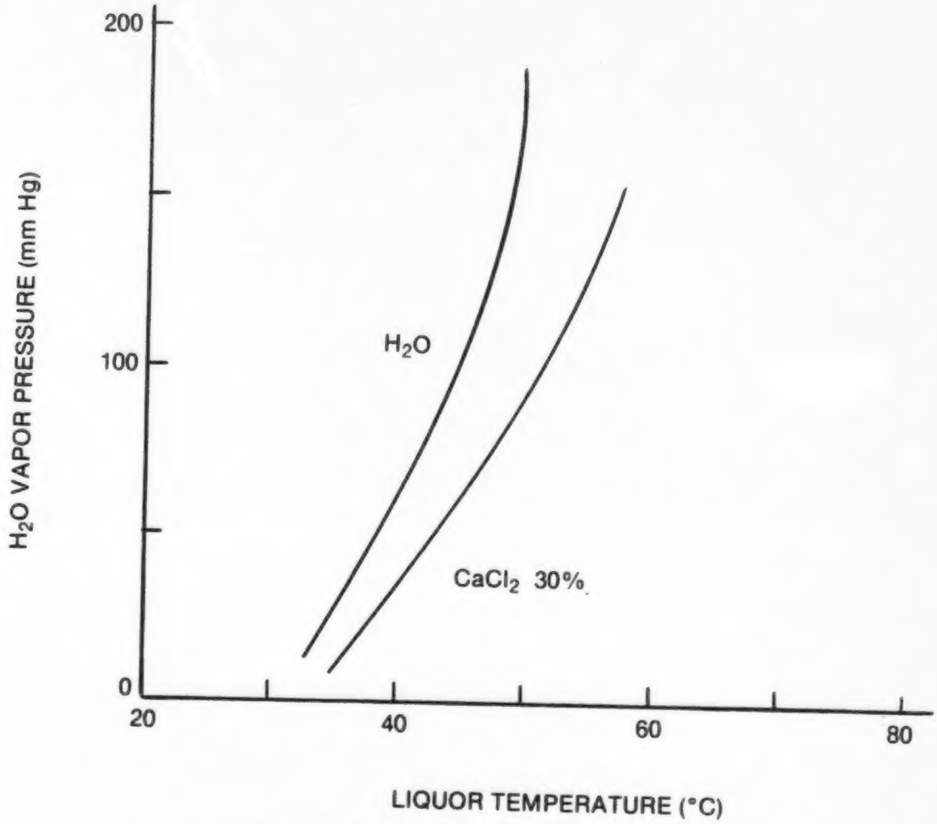


**Figure 5-11** **Vapor Pressure on $CaCl_2$ Solution.**

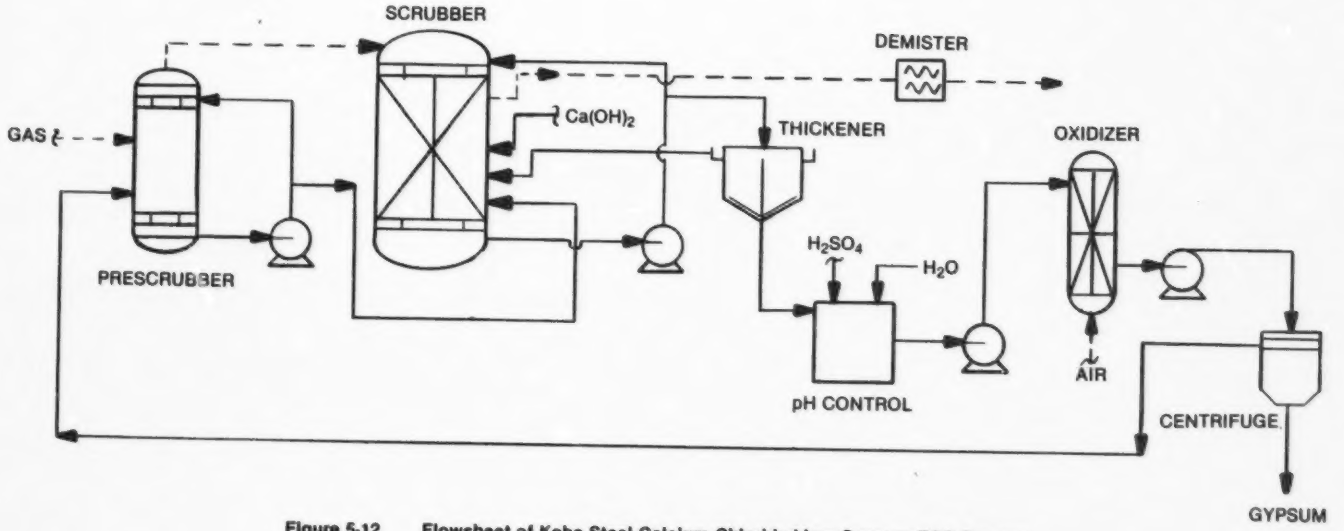

SCRUBBER
DEMISTER
GAS
$Ca(OH)_2$
THICKENER
OXIDIZER
PRESCRUBBER
$H_2SO_4$
$H_2O$
AIR
pH CONTROL
CENTRIFUGE
GYPSUM

Figure 5-12 Flowsheet of Kobe Steel Calcium Chloride-Lime-Gypsum FGD Process.

TABLE 5-11. OPERATION DATA FOR KOBE STEEL'S KAKOGAWA UNIT

| | |
|---|---|
| Flue gas treated, $Nm^3/hr$ | 720,000 (220 MW equivalent) |
| Inlet $SO_2$, ppm | 240 - 300 |
| Outlet $SO_2$, ppm | Below 30 |
| Pressure drop, mm $H_2O$ | 180 - 200 |
| Requirements | |
| Lime | 1.05 mol / mol of $SO_2$ |
| Sulfuric Acid | 0.05 mol / mol of $SO_2$ |
| $CaCl_2$ | 0.02 mol / mol of $SO_2$ |
| Electric Power, kW | 4,560 |
| Gypsum byproduct | |
| Moisture, % | 6 - 10 |
| Chloride, ppm | 100 - 200 |
| Purity, % | Over 98 |

### 5.5.3 Application to Coal-Fired Boilers

Since May 1981, Steag of West Germany, has operated a coal pilot plant to test this process with a flue gas flow rate of 5,000 $Nm^3/hr$ and flue gas concentrations of 1,000 ppm $SO_2$ and 200 ppm HCl. Over 90% of the $SO_2$ is removed using a spray tower absorber to recover 30 kg/hr of a good quality gypsum useful for cement and wallboard applications.

Since flue gas from coal contains a considerable amount of chloride, it is necessary to remove this chloride from the scrubber system. To treat flue gas from a 500 MW coal-fired boiler containing 100 ppm HCl, 1.5 t/hr of scrubber liquor containing 20% $CaCl_2$ must be purged. With the standard lime/limestone process, 60 t/hr should be purged to maintain a chloride concentration of below 5,000 ppm in the scrubber liquor (Figure 2-6 of Section 2). The wastewater loads are so small that evaporation can easily be used to obtain solids, leaving no remaining wastewater to be treated.

### 5.5.4 Evaluation

The process uses a chloride-rich liquor and thus requires corrosion-resistant materials for construction. The prescrubber also may require a high quality material. On the other hand, the process is advantageous because it requires less reheating and less water and produces virtually no wastewater. The process may suit countries such as West Germany that do not allow the purging of wastewater from FGD systems. The process may also be useful for production of throw-away gypsum without wastewater. The inlet and outlet chloride balances when the flue gas contains 2,000 ppm $SO_2$ and 120 ppm HCl and the gypsum contains about 2% $CaCl_2$ (10% liquor containing 20% $CaCl_2$).

## 5.6 SODIUM ACETATE-LIME-GYPSUM FGD PROCESS

Kureha Chemical Industries (KCI) has developed a sodium acetate-lime process which produces salable gypsum. KCI constructed a unit using this process for Toyo Rubber Co. at Sendai with a capacity of treating 40,000 $Nm^3/hr$ of flue gas from an oil-fired boiler.

A flowsheet of the KCI process is shown in Figure 5-13. Flue gas is treated in a multi-stage scrubber with a sodium acetate liquor-containing gypsum. The concentration of sodium acetate is below 1%. Over 99% of the $SO_2$ is removed at an L/G ratio of 2 $l/Nm^3$. The slurry leaving the scrubber contains sodium sulfite, acetic acid and gypsum and is treated with lime and air in a reactor to form sodium acetate and gypsum. The pH of the slurry in the reactor is about 7. A major portion of the slurry is returned to the scrubber while another portion is sent to a centrifuge where it is separated into gypsum and other components. A thickener will be installed later for other units.

As the pH is lowered by $SO_2$ absorption, a portion of acetic acid is vaporized in the lower stage of the scrubber. The volatilized acetic acid is subsequently recovered at the upper stage. Thus, over 99% $SO_2$ recovery

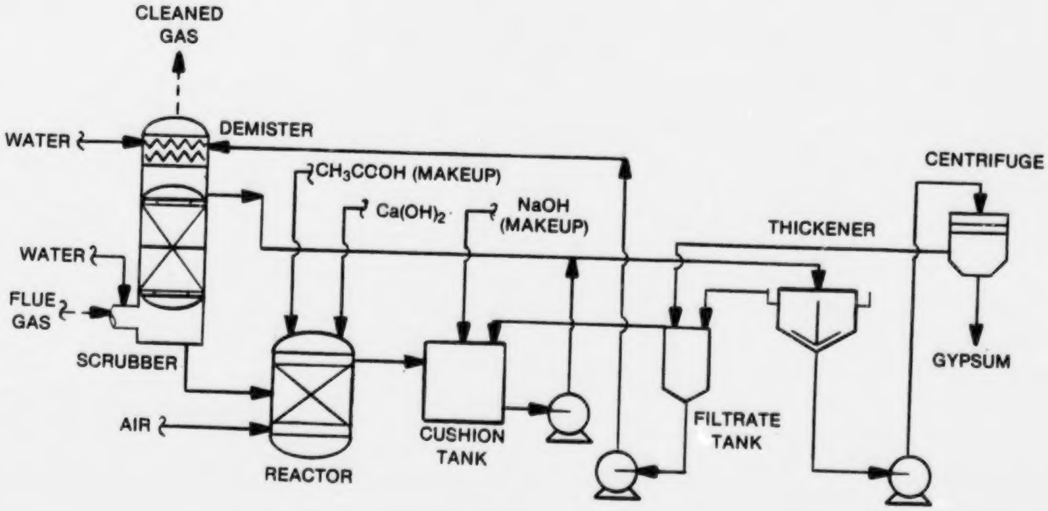

CLEANED GAS
WATER
DEMISTER
CH3CCOH (MAKEUP)
Ca(OH)2
NaOH (MAKEUP)
CENTRIFUGE
THICKENER
WATER
FLUE GAS
SCRUBBER
AIR
REACTOR
CUSHION TANK
FILTRATE TANK
GYPSUM

**Figure 5-13** **Flowsheet of the Sodium Acetate FGD Process.**

is achieved since $SO_2$ is more readily absorbed than the acetic acid vapor. Sodium sulfate does not accumulate in the scrubber liquor. Such an accumulation, which has been one of the major problems associated with conventional sodium-lime alkali FGD processes, is eliminated by the effect of the acetate.

The design parameters and operation data of the Toyo Rubber FGD unit are shown in Table 5-12. The boiler has been operated primarily at reduced loads, producing flue gas at a rate of 25,150 $Nm^3$/hr. The average reliability of the FGD unit was 98.7% in the year following its November 1980 start-up. Minor start-up problems were encountered in the first two months, but other than that, the reliability has been virtually 100%.

The power consumption of the FGD unit is low--1.4 - 1.7% of the power generated by the boiler. A small amount of acetic acid is added since a portion of the acid is decomposed or lost during the process. The gypsum by-product is of a good quality and is salable.

Kureha conducted pilot plant tests for several months with 5,000 $Nm^3$/hr of simulated coal flue gas obtained by adding fly ash and chloride to flue gas from heavy oil boilers. Since no wastewater is produced, the chloride concentration of the scrubber liquor reached about 5%, but did not affect scrubber operation. It was found that chloride reduces the decomposition of acetic acid. Kureha estimates that both investment and operation costs are less than those of the standard lime/limestone gypsum process.

The process has been licensed to Lurgi, West Germany and General Electric, U.S. Tests with actual flue gas from coal are needed to prove the usefulness of the process for coal-fired boilers.

KCI recently modified the process and has developed a sodium sulfosuccinate-limestone process to by-produce salable gypsum, which may be less expensive than the acetate-lime process.

TABLE 5-12. OPERATION PARAMETERS OF THE TOYO RUBBER FGD UNIT

| | | Design | Actual | Actual |
|---|---|---|---|---|
| Flue gas treated, $Nm^3/hr$ | | 40,000 | 40,000 | 25,150 |
| Flue gas temperature, °C | Inlet | 220 | 170 | 170 |
| | Outlet | 60 | 58 | 58 |
| $SO_2$, ppm | Inlet | 1,350 | 1,270 | 1,270 |
| | Outlet | below 5 | 0.5 | 0.5 |
| $SO_2$ removal efficiency, % | | 99.6 | 99.96 | 99.96 |
| Particulate, $mg/Nm^3$ | Inlet | below 200 | 100 | 100 |
| | Outlet | below 100 | 10 | 10 |
| Pressure drop, mm $H_2O$ | | 221 | 220 | 125 |
| Consumptions | | | | |
| $Ca(OH)_2$, kg/hr | | 185 | 163 | 102 |
| $CH_3COOH$, kg/hr | | 9 | 3.3 | 2.1 |
| Water, ton/hr | | 6 | 2.9 | 1.8 |
| Power, kW | | 220 | 167 | 142 |
| Gypsum byproduct | | | | |
| Production, kg/hr | | 420 | 381 | 244 |
| Purity, % | | over 95 | 97.4 | 97.4 |
| Moisture, % | | below 10 | 8 | 8 |
| System reliability, % | | over 95 | - | 98.7 |

## 5.7 RED MUD DESULFURIZATION SYSTEM (RMDS)

### 5.7.1 Development Status

Sumitomo Aluminum Smelting Co. has developed an FGD process using red mud, a waste byproduct of aluminum production from bauxite, as the absorbent. In 1977, Sumitomo Chemical Engineering Co. constructed a commercial RMDS unit with a capacity of treating 220,000 $Nm^3/hr$ of flue gas from an oil-fired boiler at Niihama, Sumitomo Joint Electric Power. Sumitomo Joint Electric Power is planning to construct a coal-fired boiler, to which the RMDS process also will be applied.

The typical chemical composition of red mud is shown in Table 5-13. The mud contains about 9% sodium and therefore is useful for $SO_2$ removal. A few companies studied the use of the mud for FGD and experienced several problems. The major problems have been: 1) low reactivity of the sodium present in the mud in the form of sodalite, ($Na_2O \cdot Al_2O_3 \cdot 2SiO_2 \cdot 1/3Na_2X$) and 2) scale formation in the scrubber system due to red mud and gypsum which is formed by the reaction of calcium carbonate in the mud with $SO_2$ and $O_2$. Sumitomo Aluminum Smelting Co. has solved the first problem by using a two stage absorber and air oxidation system and the second problem with pH control and a special washing system.

TABLE 5-13. TYPICAL COMPOSITION OF RED MUD

| | |
|---|---|
| $Na_2O$ | 9% |
| $Al_2O_3$ | 22% |
| $SiO_2$ | 17% |
| $Fe_2O_3$ | 32% |
| $TiO_2$ | 6% |
| $CaCO_3$ | 3% |

### 5.7.2 Process Description

A flowsheet of the commercial unit is shown in Figure 5-14. The boiler flue gas is first cooled by water spray to 60°C and then introduced into a two stage absorber. The red mud slurry, with a concentration of 300 g/l, is fed through the No. 1 tank to the upper stage (2nd stage) of the absorber. The partially reacted slurry is then fed through the No. 2 tank to the bottom stage (1st stage) of the absorber for further $SO_2$ removal. The reaction forms $NaHSO_3$ and $Na_2SO_3$. Air is blown into the No. 1 and No. 2 tanks to oxidize part of the compounds to $NaHSO_4$ and $Na_2SO_4$. It is important to maintain the oxidation ratio at a suitable level in order to promote $SO_2$ removal as well as to prevent scaling.

The slurry leaving the No. 2 tank is led into the oxidizing tank for further oxidation which eliminates some of the COD. The resulting slurry is discarded.

### 5.7.3 Performance and Economics

The RMDS unit has been operated continuously since 1977 while the boiler has been in operation. The boiler undergoes scheduled maintenance

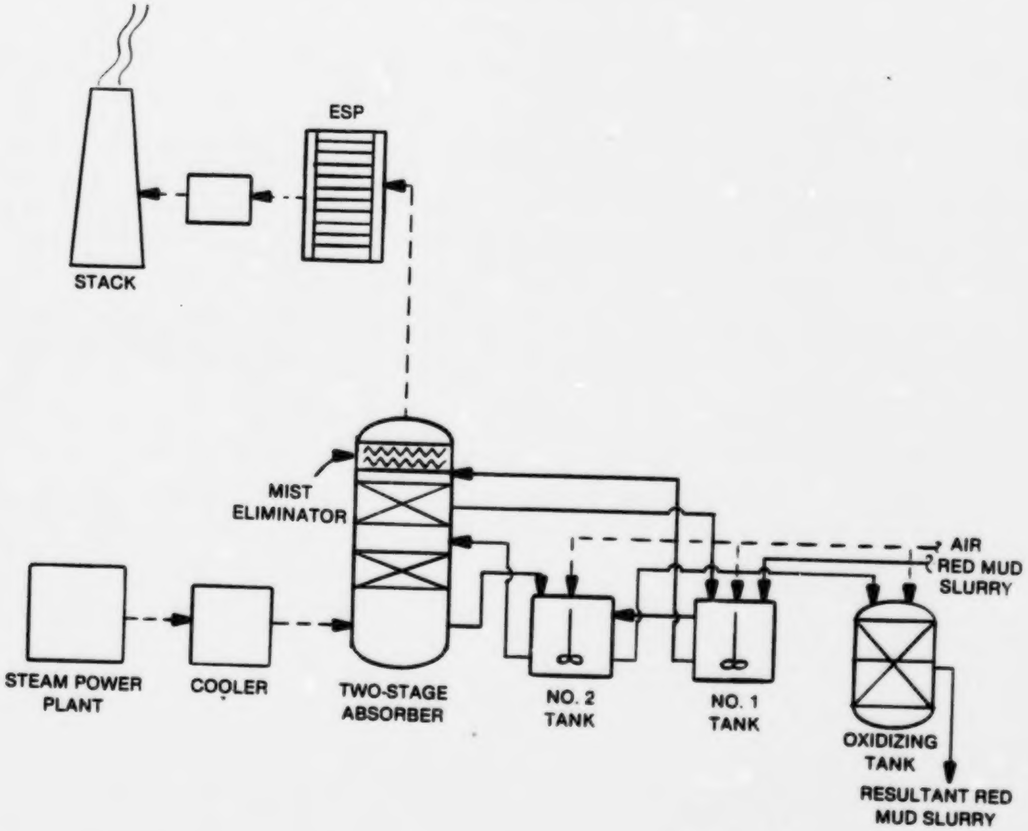

ESP
STACK
MIST ELIMINATOR
AIR
RED MUD SLURRY
STEAM POWER PLANT
COOLER
TWO-STAGE ABSORBER
NO. 2 TANK
NO. 1 TANK
OXIDIZING TANK
RESULTANT RED MUD SLURRY

Figure 5-14 Flowsheet of the Red Mud Desulfurization Unit.

once a year. Over 70% of the sodium in the mud reacts to remove over 95% of the $SO_2$ in the flue gas. Examples of operation data are given in Table 5-14. About 70% of the dust in the flue gas is also removed by scrubbing and red mud mists are entirely removed by mist eliminators. Table 5-15 compares the economics of the red mud process with those of the limestone-gypsum process. As indicated, the red mud process is considerably less costly than the limestone scrubbing process.

### 5.7.4 Evaluation

Using aluminum plant waste mud as the absorbent, the RMDS process has successfully removed over 95% of flue gas $SO_2$. For power plants located near aluminum plants which produce red mud, the process may be useful because of its simplicity and low cost. The process may be particularly suitable for coal-fired boilers since fly ash from coal may improve the physical properties of the red mud waste.

TABLE 5-14. OPERATION RESULTS OF RMDS PLANT

| | Maximum Load | Minimum Load |
|---|---|---|
| Flow rate waste gas, $Nm^3/hr$ | 220,000 | 100,000 |
| Inlet $SO_2$ concentration in waste gas, ppm | 1,260 | 1,040 |
| Outlet $SO_2$ concentration, ppm | 55 | 17 |
| Efficiency of $SO_2$ removal, % | 96 | 98 |
| Inlet dust concentration in waste gas, $mg/Nm^3$ | 165 | 178 |
| Outlet dust concentration, $mg/Nm^3$ | 53 | 46 |
| Reacted NaOH in sodalite, % | 73 | 73 |

TABLE 5-15. COMPARISON OF ECONOMIC DATA ON RMDS AND LIMESTONE-GYPSUM SCRUBBING PROCESSES (Yen)

| | Limestone-Gypsum Scrubbing Plant | RMDS Plant |
|---|---|---|
| Construction, yen | 100 | 65 |
| Operating Cost, yen | 100 | 70 |

# SECTION 6

## $SO_x$ AND $NO_x$ REMOVAL

### 6.1 INTRODUCTION

The combination of selective catalytic reduction (SCR) of $NO_x$ with wet process FGD has been used commercially for several coal-fired boilers since 1980. During the early stages of SCR development (1975-76), the SCR reactor was placed downstream of the FGD system to protect the SCR catalyst from erosion by fly ash and also from $SO_x$ poisoning. The SCR system, however, requires a large amount of fuel for gas heating. Since parallel-flow catalysts free from both dust plugging and $SO_x$ attack have been developed, new plants place the SCR reactor between the boiler and the 350-400°C air preheater. The FGD unit is placed downstream of the SCR and air preheater (Sections 4.2 and 4.3).

There are two SCR systems available for treatment of flue gas from coal-fired boilers. One is the "high-dust" system which treats the flue gas with full dust loads; the other is the "low-dust" system which has a hot-side ESP upstream of the SCR reactor to remove fly ash (Section 2.7.2). The hot-side ESP is expensive but is suitable for low-sulfur coals for which cold-side ESP is not highly efficient. Extensive tests comparing the two systems have been conducted at the Nakoso Station of Joban Joint Power.

Tests on dry simultaneous removal of $SO_x$ and $NO_x$ also have been conducted. The electron beam process for simultaneous removal has been tested by the Ebara Manufacturing Co. mainly for flue gases from oil-fired boilers and iron-ore sintering machines. In the U.S., AVCO, the licensee of the Ebara process, has conducted tests on the effects of coal fly ash on the electron beam process.

The activated carbon process for the simultaneous removal of $SO_x$ and $NO_x$ was tested by EPDC jointly with Sumitomo Heavy Industries. A demonstration plant will be completed by 1984. Activated carbon processes also have been tested by Mitsui Mining Co.

Wet simultaneous $SO_x/NO_x$ removal processes have been tested and applied to several small commercial units mainly for oil-fired boilers. The wet processes, however, may not be suitable for large scale applications, particularly for coal-fired boilers. Large scale systems need wastewater treatment to remove a considerable amount of impurities including nitrate and nitrite (3).

## 6.2 SCR-FGD COMBINATION AT NAKOSO PLANT, JOBAN JOINT POWER (17)

Tokyo Electric Power Co., jointly with Tohoku Electric Power Co., Joban Joint Power Co., and MHI, has carried out extensive pilot plant tests at Nakoso Station on combined systems of SCR (high-dust and low-dust) with wet limestone-gypsum process FGD using 4,000 $Nm^3/hr$ of flue gas from a coal-fired boiler. A flowsheet of the pilot plant is shown in Figure 6-1. The high-dust SCR reactor treats flue gas with about 20 $g/Nm^3$ of dust while the reactor of low-dust system treats flue gas containing 0.1-0.2 $g/Nm^3$ of dust. In 1979, the high-dust system was operated for 5,000 hours and the low-dust system for 4,000 hours. Further tests were conducted in 1980.

In both systems a honeycomb catalyst with a gas down-flow configuration is used to remove 80-90% of the $NO_x$. With the high-dust system, erosion of the catalyst by the fly ash can be prevented by placing a ceramic "dummy spacer", identical in shape to the catalyst, on top of the honeycomb catalyst layer. Ammonium bisulfate deposition in the air preheater, the major problem with SCR, can be prevented by maintaining the $NH_3$ concentration below 5 ppm at the reactor outlet and by soot-blowing once a day. Ammonium bisulfate did not deposit noticeably after these procedures were implemented because of the cleaning effect of the fly ash.

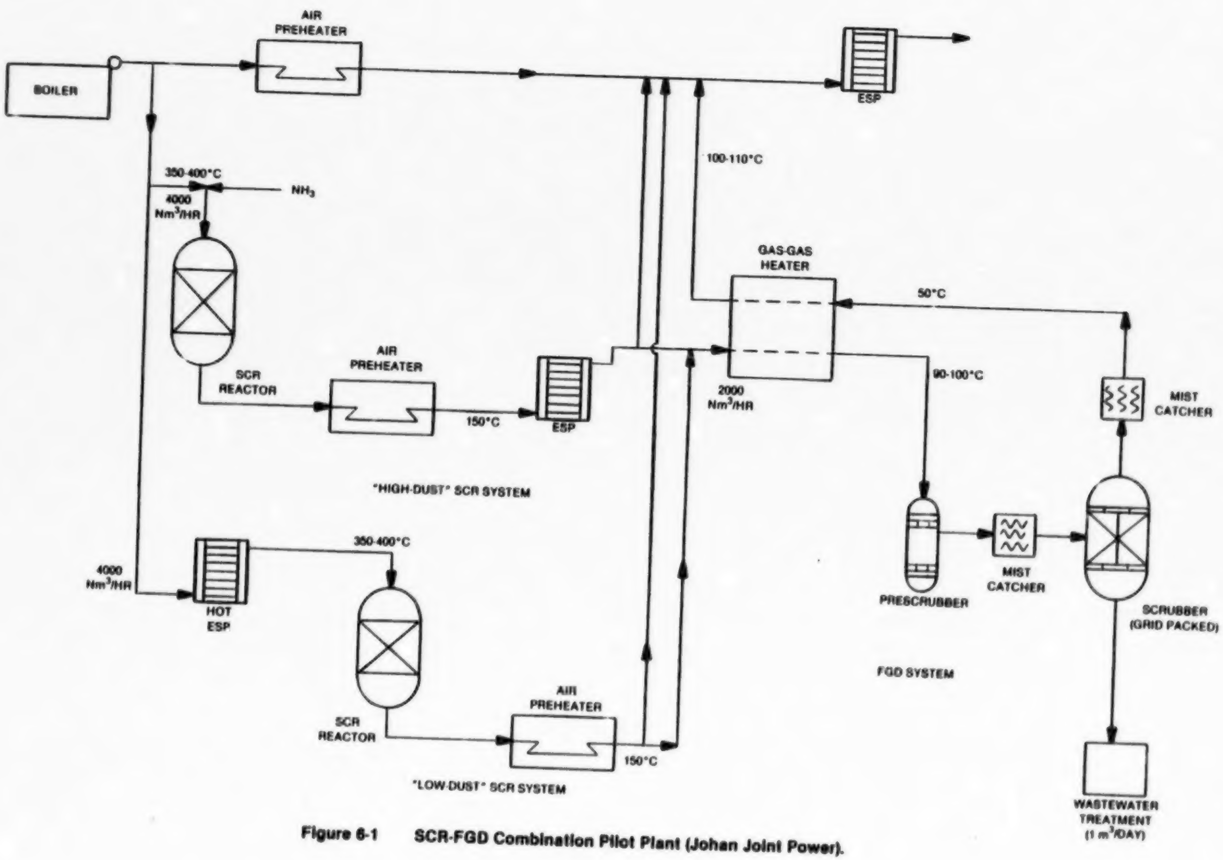

BOILER
AIR PREHEATER
ESP
100-110°C
350-400°C
NH$_3$
4000 Nm$^3$/HR
SCR REACTOR
AIR PREHEATER
150°C
ESP
GAS-GAS HEATER
50°C
2000 Nm$^3$/HR
90-100°C
MIST CATCHER
"HIGH-DUST" SCR SYSTEM
4000 Nm$^3$/HR
HOT ESP
350-400°C
SCR REACTOR
AIR PREHEATER
150°C
"LOW-DUST" SCR SYSTEM
PRESCRUBBER
MIST CATCHER
SCRUBBER (GRID PACKED)
FGD SYSTEM
WASTEWATER TREATMENT (1 m$^3$/DAY)

Figure 6-1 SCR-FGD Combination Pilot Plant (Johan Joint Power).

On the other hand, with the low-dust system, the small amount of dust leaving the hot ESP consists of very fine particles which are rather sticky and tend to deposit at the inlet of the honeycomb. Also, the air preheater requires soot-blowing 3 times a day to prevent ammonium bisulfate deposition.

For both systems, the FGD unit successfully removed over 90% of the $SO_2$. The FGD unit was similar to that at Shimonoseki, Chugoku Electric (Section 4.2). A semi-venturi spray scrubber developed by MHI was used for prescrubbing to remove most of the dust, chloride, fluoride, etc. in the flue gas. Tests indicated that the dust contained in the flue gas in concentrations of 100, 200, and 300 mg/$Nm^3$ was reduced to about 20, 30, and 40 mg, respectively, by the prescrubber and to about 15, 20, and 30 mg, respectively by the scrubber with plastic grid packings (Figure 6-2).

It was concluded that both high- and low-dust SCR systems and their combination with FGD worked well and could be used commercially. Joban Joint Power has begun construction of 2 new boilers, each with a capacity of 600 MW, which initially will use oil and coal. Both boilers will use high-dust SCR with a honeycomb catalyst and lime-stone-gypsum FGD. The SCR unit for one of the boilers is to be constructed by MHI; IHI will construct the unit for the other boiler. FGD units for both boilers will be constructed by MHI.

## 6.3 EBARA ELECTRON BEAM PROCESS

### 6.3.1 Introduction

Ebara Corporation, jointly with the Japan Atomic Energy Research Institute, has developed a process which simultaneously removes $SO_x$ and $NO_x$ in flue gas by electron beam irradiation; the resulting particulates are collected with an electrostatic precipitator. An electric beam process pilot plant with a capacity of treating 1,000 $Nm^3$/hr of flue gas from an oil-fired boiler was operated by Ebara from 1974 through 1977. Tests during the early stage of development were not very successful. Later Ebara began using ammonia, which substantially improved the process.

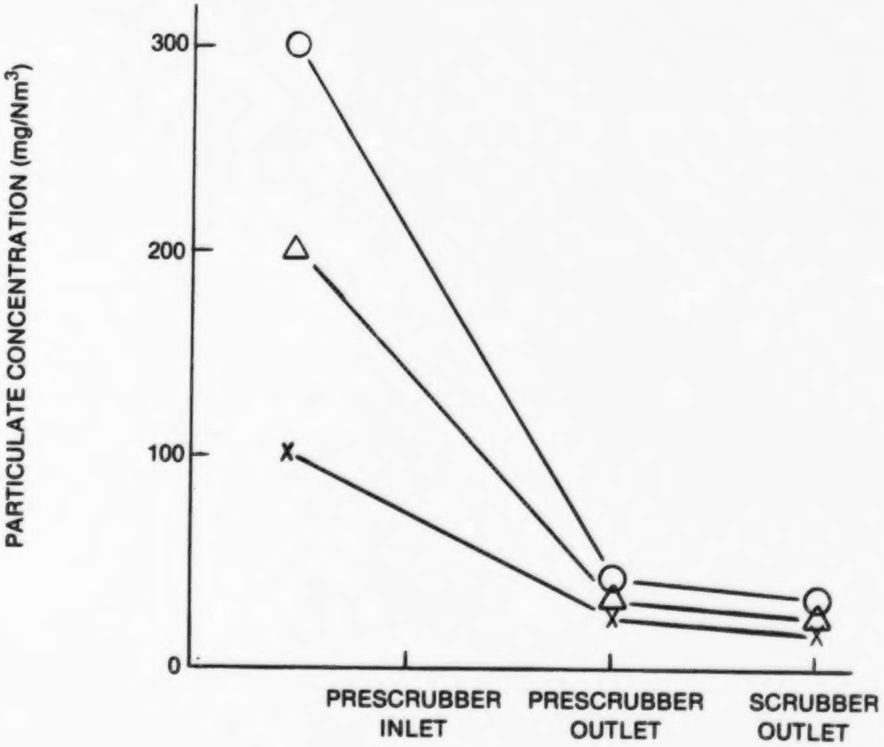


**Figure 6-2** **Particulate Removal by FGD System (Nakoso).**

70A2521

By irradiating the gas-containing ammonia, over 90% of the $SO_x$ and over 80% of the $NO_x$ can be removed, thereby forming fine crystals of ammonium sulfate nitrate (double salt) which are useful for fertilizer. A larger pilot plant with a capacity of treating 3,000 to 10,000 $Nm^3$/hr of flue gas from an iron-ore sintering machine was operated from July 1977 through June 1978 at Nippon Steel's Yawata Works. The process has been licensed to AVCO, USA which has conducted tests on the effects of fly ash from coal-fired boilers. Recently, the U. S. Department of Energy (DOE) signed a contract with AVCO and Ebara for further study of the process (18).

### 6.3.2 Pilot Plant Tests (19,20)

A flowsheet of the larger electron beam process pilot plant is shown in Figure 6-3. Major equipment of the plant includes gas coolers which use indirect cooling (heat exchanger) and direct cooling (water spray), ammonia injection facilities, an electron beam reactor surrounded by a concrete wall to prevent x-ray emissions, an ESP, and a control system. As shown in Figure 6-3, two electron beam accelerators, with a unit capacity of 750 kV, 60 mA each were placed at opposite sides of a cylindrical reactor with a diameter of 2.6 m. The distribution of the dose rate in the reactor is shown in Figure 6-4.

Since the dose rate is not uniform, an impeller was placed at the inlet of the reactor to rotate the gas and homogenize the reaction. A side stream of the gas from an iron-ore sintering machine was cooled to 70-90°C, injected with ammonia, and introduced into the reactor. The stream contained about 200 ppm each of $SO_2$ and $NO_x$ (mainly NO), 40 mg/$Nm^3$ of dust, 15.5% $O_2$, 10.3% $H_2O$, and 16.2% $CO_2$, at a rate of 3,000 - 10,000 $Nm^3$/hr. In some of the tests, $SO_2$, NO, and fly ash were added to the flue gas to simulate coal flue gas. Ammonia normally was used at a stoichiometry, $RNH_3$, of 1.0 (2 mol $NH_3$ to 1 mol $SO_x$ and 1 mol $NH_3$ to 1 mol $NO_x$). In some of the tests, the stoichiometry was changed.

The relationship between the gas rotation ratio and the removal efficiency is shown in Figure 6-5. A maximum efficiency of over 95% for $SO_x$ and

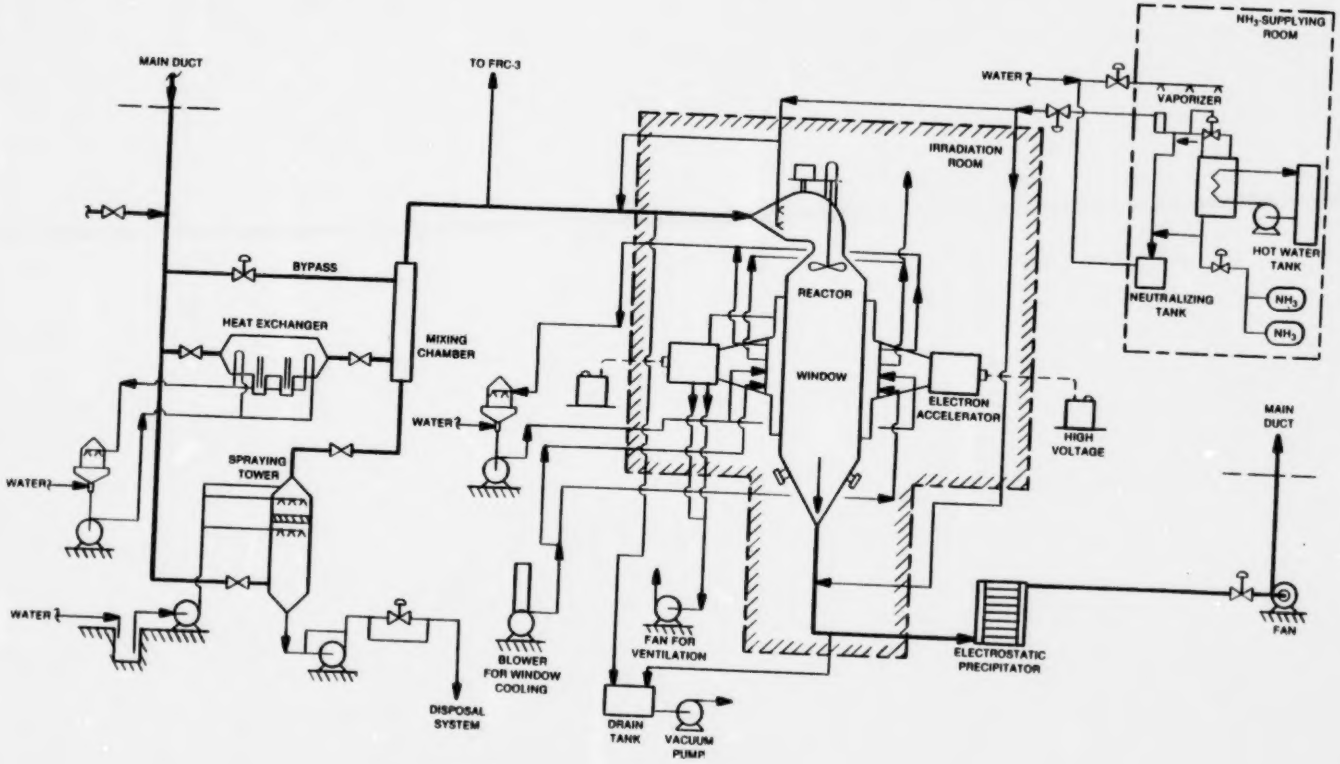

MAIN DUCT
TO FRC-3
WATER
NH$_3$-SUPPLYING ROOM
VAPORIZER
IRRADIATION ROOM
BYPASS
HEAT EXCHANGER
MIXING CHAMBER
REACTOR
WINDOW
HOT WATER TANK
NEUTRALIZING TANK
NH$_3$
NH$_3$
ELECTRON ACCELERATOR
HIGH VOLTAGE
MAIN DUCT
WATER
WATER
SPRAYING TOWER
WATER
FAN FOR VENTILATION
BLOWER FOR WINDOW COOLING
DISPOSAL SYSTEM
DRAIN TANK
VACUUM PUMP
ELECTROSTATIC PRECIPITATOR
FAN

Figure 6-3 Flowsheet of Electron Beam Process Pilot Plant (Yawata Works).

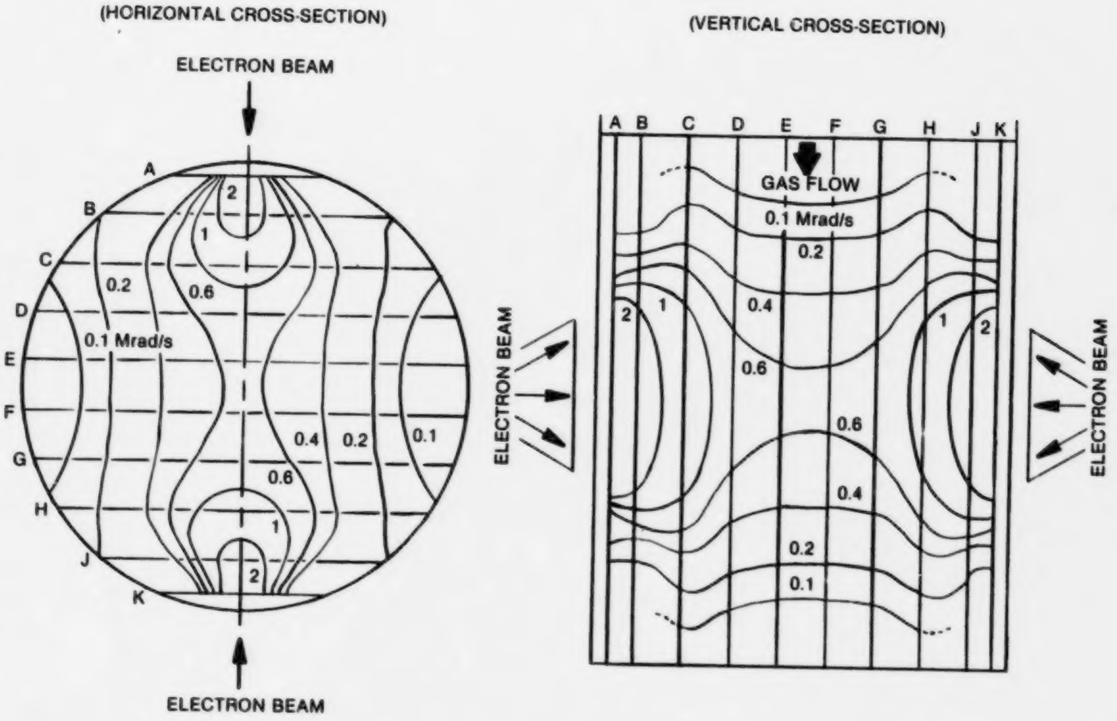

(HORIZONTAL CROSS-SECTION)
ELECTRON BEAM
A
B
C
D
E
F
G
H
J
K
2
1
0.2
0.6
0.1 Mrad/s
0.4
0.2
0.1
0.6
1
2
ELECTRON BEAM
(VERTICAL CROSS-SECTION)
A B C D E F G H J K
GAS FLOW
0.1 Mrad/s
0.2
0.4
2
1
0.6
1
2
0.6
0.4
0.2
0.1
ELECTRON BEAM
ELECTRON BEAM

**Figure 6-4** Dose Rate Distribution in Reactor (accelerating voltage = 600 kV).

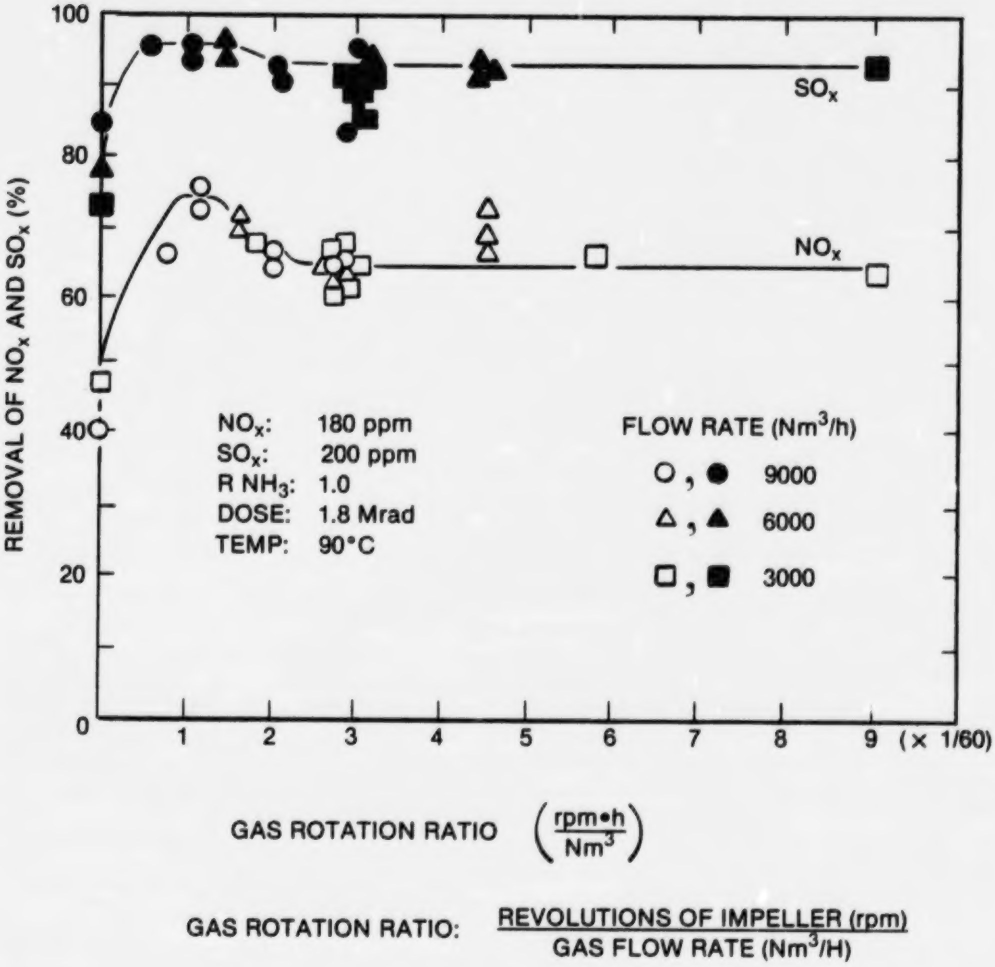


**Figure 6-5** Relationship between Gas Rotation Ratio and Removal Efficiency.

about 80% for $NO_x$ was obtained at the ratio of about 1/60 with 1.8 Mrad dose at 90°C.

The effect of the dose on the efficiency and $NH_3$ leakage is shown in Figure 6-6. The $SO_x$ removal efficiency exceeded 90% at 1.3 Mrad at 70°C and at 2 Mrad at 90°C, while $NO_x$ removal efficiency reached a maximum of about 80% with 1.8 Mrad at 70°C or about 65% with 1.8 Mrad at 90°C. The decrease in $NO_x$ removal efficiency at higher doses may be due to the formation of $NO_x$ from $N_2$ and $O_2$. $NH_3$ leakage decreased with increasing doses and was almost zero at 70°C above 1.8 Mrad. Leakage was considerable at 90°C even with a 2.4 Mrad dosage. The tests shown in Figure 6-6 were conducted using a gas rotation ratio of 1/33. Better results may be obtained with a ratio of 1/60.

Figure 6-7 shows the results of 600 hours operation of the pilot plant at 1.5 Mrad and 90°C with the rotation ratio of 1/33. During the 600 hours, the test was stopped twice due to the shutdown of the sintering machine. The inlet $SO_x$ and $NO_x$ concentrations ranged from 180 to 230 ppm and 170 to 210 ppm, respectively. The $SO_x$ and $NO_x$ removal efficiencies ranged from 92 to 97% and 75 to 85%, respectively, while the $NH_3$ concentration at the reactor outlet ranged from 10 to 50 ppm. Considerable ammonia leakage was caused by frequent fluctuation in the inlet $SO_x$ and $NO_x$ concentrations as well as by problems with the ammonia flow meter.

The balance of $NO_x$, $NH_3$, and $SO_x$ in 3 runs is shown in Table 6-1. Most of the $NO_x$, $NH_3$, and $SO_x$ were reacted and caught by the ESP, but a considerable portion of $NH_3$ and $SO_x$ was deposited in the duct and a large portion of $NO_x$ was emitted from the recovery system. The totals of $NH_3$ and $SO_x$ were a little larger than 100%, presumably due to sampling and analysis errors. The

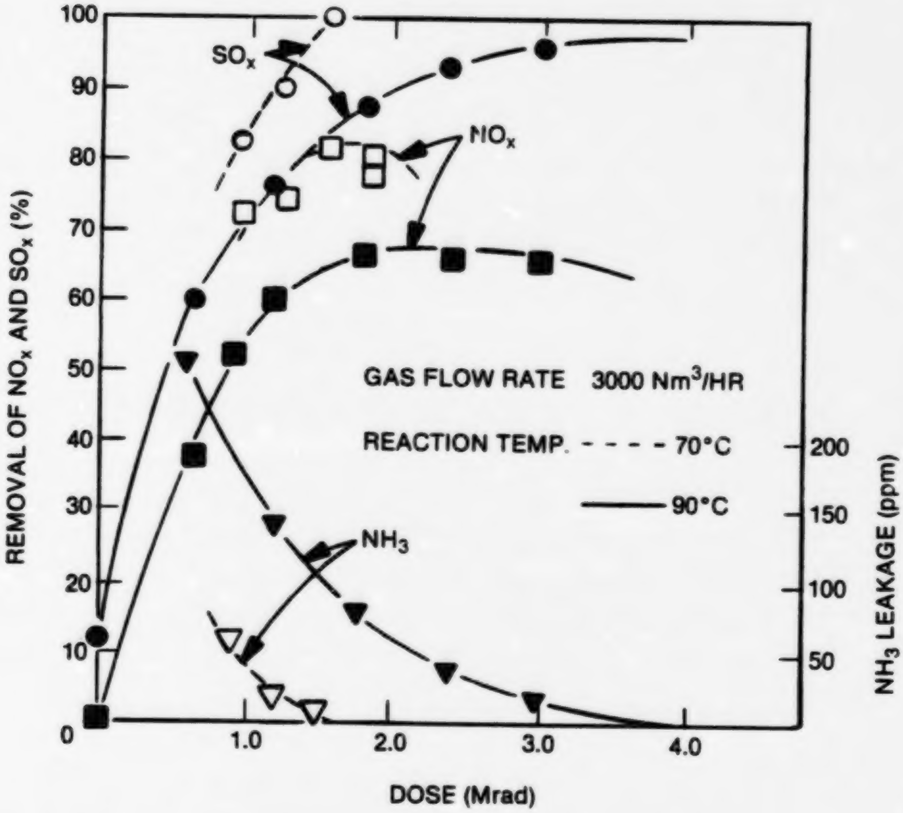


Figure 6-6 Effect of Dose on Removal Efficiency and $NH_3$ Leakage (Gas Rotation Ratio: 1/33).

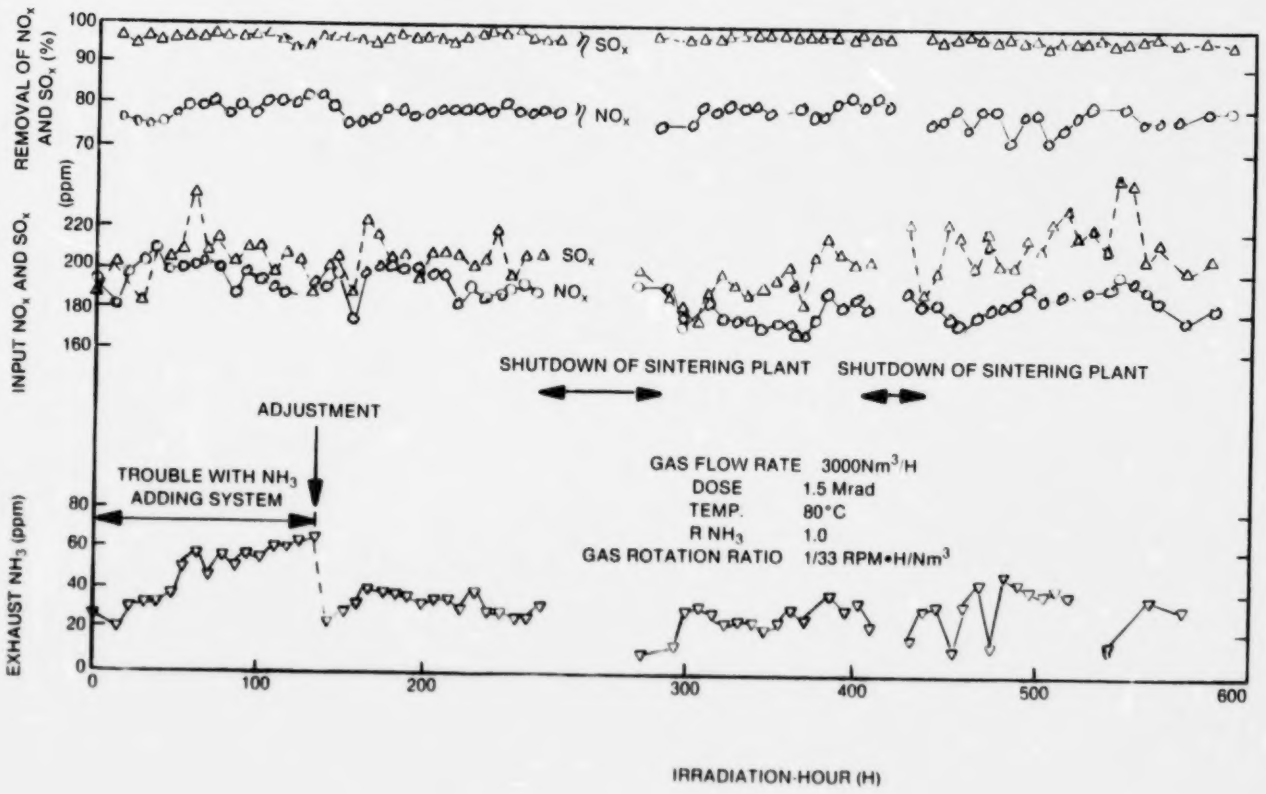


**Figure 6-7** Results of Continuous Test of Ebara Process Pilot Plant

TABLE 6-1. BALANCE OF $NO_x$, $NH_3$ AND $SO_x$ (PERCENT OF INLET)

| Run No. | Component | Caught by ESP | Deposit in duct | Emitted | Total |
|---|---|---|---|---|---|
| 1109 | $NO_x$ | 81 | 6 | 49 | 136 |
| | $NH_3$ | 64 | 36 | 9 | 109 |
| | $SO_x$ | 62 | 43 | 10 | 115 |
| 1111 | $NO_x$ | 87 | 6 | 31 | 124 |
| | $NH_3$ | 65 | 36 | 10 | 111 |
| | $SO_x$ | 64 | 40 | 8 | 112 |
| 1114 | $NO_x$ | 78 | 6 | 38 | 132 |
| | $NH_3$ | 65 | 36 | 7 | 105 |
| | $SO_x$ | 59 | 43 | 8 | 110 |

$NO_x$ totals were also considerably larger than 100%, indicating the formation of $NO_x$ from $N_2$ and $O_2$ caused by the irradiation. Chemical analysis of the exhaust gas showed the presence of $N_2O$ in a concentration of 10 to 20 ppm.

The titanium foil at the accelerator window was often broken during pilot plant testing. Later, a foil with a titanium alloy was used which lasted for over 2 months. Ebara now has developed a way to change the foil without stopping the accelerator operation.

An x-ray diffraction analysis of products caught by the ESP showed that the major compounds were $2NH_4NO_3 \cdot (NH_4)_2SO_4$, $3NH_4NO_3 \cdot (NH_4)_2SO_4$ and $(NH_4)_2SO_4$. Chemical analysis of a product obtained from gas containing 220 ppm $SO_2$ and 235 ppm $NO_x$ with 700 ppm $NH_3$ at 90°C indicated that the product contained 33.6% $SO_4$, 29.4% $NO_3$, 23.6% $NH_3$, and 0.9% C.

The results of tests with $SO_x$- and $NO_x$-rich flue gas are shown in Figure 6-8. About 80% of both $SO_x$ and $NO_x$ were removed at 1.8 Mrad with an ammonia stoichiometry of 1.0, which produced an ammonia leakage of about 50 ppm. The gas rotation was poor (1/17) because the ammonia supply was limited and the flue gas volume was only 1,500 $Nm^3$/hr. It is expected that

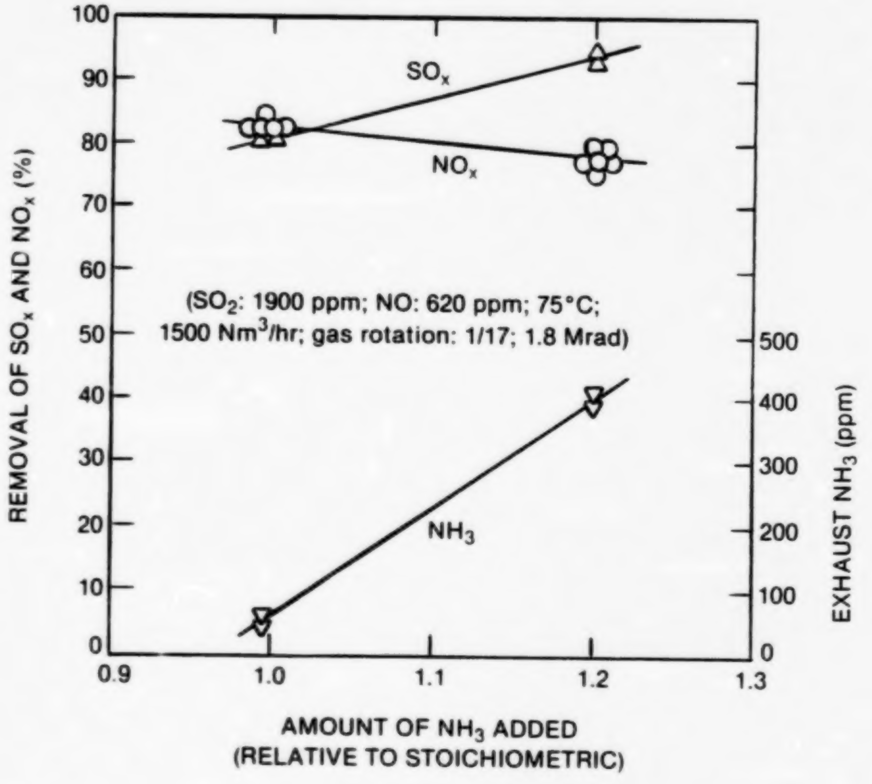


**Figure 6-8** Removal of $SO_x$ and $NO_x$ vs. Amount of Ammonia Added.

a 90% removal rate for both $SO_x$ and $NO_x$ may be attained with a better rotation as shown in Figure 6-5.

Tests of fly ash additions of 0.1 - 0.5 g/$Nm^3$ (equivalent to 95 - 75% removal of fly ash from pulverized coal) indicated that the fly ash had no adverse effect on $SO_x$ and $NO_x$ removal efficiencies (18).

### 6.3.3 Reaction Mechanism

Ebara also conducted fundamental tests on the reaction mechanism which takes place in this system. Although the reactions are complex, the major one seems to be the formation of oxidant radicals such as $OH^-$ by the radiation of water vapor, which oxidizes $SO_2$ and NO to form sulfuric acid and nitric acid mists. The mists then react with ammonia to form fine crystals of sulfate and nitrate. Tests also have shown that the $SO_2$ removal efficiency is slightly higher when the gas is cooled by water spray than when cooled by heat exchanger. The $NO_x$ removal efficiency was not appreciably affected by the type of cooling mechanism. Presumably the fine water spray droplets combine with $SO_3$ to form sulfuric acid mists and thus promote the formation of $SO_3$ from $SO_2$.

### 6.3.4 Commercial Plant Assumptions

Electron beam accelerators with a capacity of 100 - 150 kW have been used commercially for various purposes without problems. A larger accelerator with a capacity of 500 kW will be commercialized in the near future. A 500 kW accelerator can treat 60,000 to 70,000 $Nm^3$/hr of flue gas. Since it is preferable to use at least four accelerators in a reactor, an economical minimum capacity for a reactor would be 240,000 to 280,000 $Nm^3$/hr, which is equivalent to 70 to 100 MW.

Figure 6-9 shows a layout plan for a commercial plant with a capacity of treating 1,000,000 $Nm^3$/hr of flue gas from a utility boiler (290 MW for coal and 350 MW for oil). The gas contains 600 ppm $NO_x$, 1,000 ppm $SO_x$, and 100 mg/ $Nm^3$ of dust. The 150°C flue gas is cooled by a heat exchanger

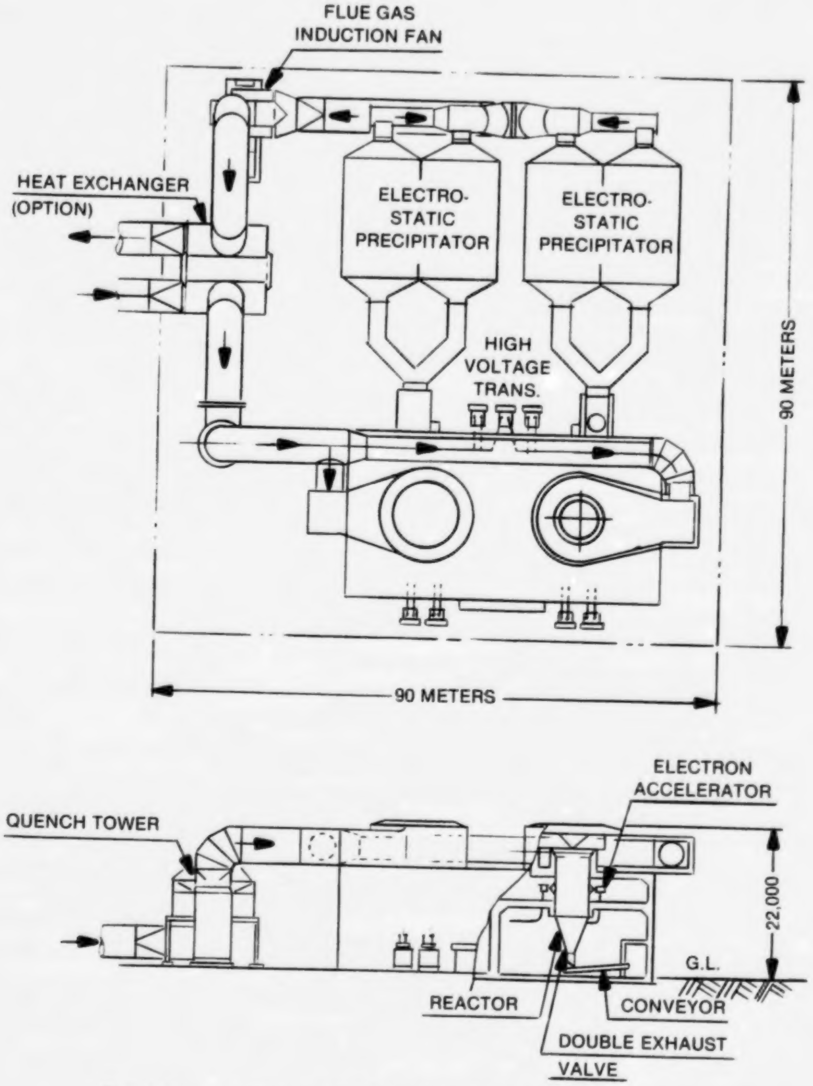

FLUE GAS
INDUCTION FAN
HEAT EXCHANGER
(OPTION)
ELECTRO-
STATIC
PRECIPITATOR
ELECTRO-
STATIC
PRECIPITATOR
HIGH
VOLTAGE
TRANS.
90 METERS
90 METERS
ELECTRON
ACCELERATOR
QUENCH TOWER
22,000
G.L.
REACTOR
CONVEYOR
DOUBLE EXHAUST
VALVE

**Figure 6-9** Layout Plan for Ebara Electron Beam Process Commercial Plant ($1,000,000\ Nm^3/hr$).

to about 110°C and then to 70°C by indirect cooling with water. The cooled gas is mixed with ammonia and treated by two reactors in parallel with a diameter of about 4 m, passed through an ESP, and then heated to 110°C by the heat exchanger.

A total of 14-16 accelerators (500 kW each) may be used to remove 80% of the $NO_x$ and 90% of the $SO_x$. The treated gas may contain 120 ppm $NO_x$, 100 ppm $SO_x$, and less than 20 ppm $NH_3$ with about 15 mg/$Nm^3$ of dust.

The investment cost for the electron beam unit was estimated by Ebara to be close to that of a conventional wet FGD unit, provided that a large accelerator (500 kW) is available at a reasonable cost. Estimated utility requirements for the 1,000,000 $NM^3$/hr plant are shown in Table 6-2. The total power consumption of 10,000 kWhr accounts for 2.9 to 3.4% of the power generated by the power plant. The power consumption approaches that of the combination of SCR of $NO_x$ and wet FGD. SCR requires 0.1 to 0.2% of the power and FGD consumes 1.6 to 2.5% excluding gas reheating.

TABLE 6-2. ESTIMATED UTILITY REQUIREMENT PER HOUR FOR EBARA ELECTRON BEAM PROCESS COMMERCIAL PLANT (Flue gas 1,000,000 $Nm^3$/hr, 350 MW for oil, 290 MW for coal)

| | | |
|---|---|---|
| Power | Electron beam accelerator<br>Others<br>Total | 6,900 kWhr<br>3,100 kWhr<br>10,000 kWhr |
| Steam | Ammonia vaporization<br>Soot blowing<br>Total | 1 tons<br>1 tons<br>2 tons |
| Water | Gas cooling<br>Other<br>Total | 33 tons<br>1 tons<br>34 tons |
| Ammonia | 0.94 of stoichiometric amount | 2 tons |

The cost for the electron beam process estimated by AVCO is compared with the cost of lime scrubbing in Table 6-3. The cost of the AVCO/Ebara process assumes the use of a simple water spray for gas cooling and is considerably less costly than lime scrubbing.

TABLE 6-3. COST COMPARISON OF FLUE GAS TREATMENT PROCESSES[c] (500 MWe) (20)

| | | Avco/Ebara | Scrubber |
|---|---|---|---|
| Total capital cost | $M | 43 | 60 |
| | $/kW | 86 | 120 |
| | | ($/MWh) | ($/MWh) |
| Depreciation and overhead[a] | | 2.4 | 3.4 |
| Variable operating cost[b] | | 2.5 | 2.8 |
| Reheat cost | | 0 | 1.0 |
| Total operating cost | | 4.9 | 7.2 |
| Byproduct charge/(credit) | | (0.6) | 0.4 |
| Net operating cost | | 4.3 | 7.6 |
| Total first year cost | | $14M | $26M |

[a] 80% load factor; annual cost = 20% of total capital.

[b] Ammonia at $130/ton. Sold as agricultural fertilizer at $15/ton. Does not include cost of disposal site and preparation.

[c] Cost estimates established by Gibbs & Hill, Inc., New York. $SO_2$ = 1900 ppm, 90% removed; $NO_x$ = 620 ppm, 90% removed; lime scrubber does not remove $NO_x$.)

### 6.3.5 Evaluation

The electron beam process is unique and has the following advantages and disadvantages:

Advantages:

(1) $SO_x$ and $NO_x$ are removed simultaneously.

(2) Because of the dry process, wastewater treatment is not needed.

(3) Ammonia, $SO_x$ and $NO_x$ are recovered as useful products.

(4) The process is simple and the operation seems easy and free from troubles except for the occasional renewal of the foil at the accelerator window.

Disadvantages:

(1) For flue gas from coal, a dust removal facility is needed upstream of the reactor to obtain a useful by-product.

(2) Gas cooling to about 70°C is needed to obtain a high removal efficiency.

(3) Appreciable amounts of $NH_3$ and $N_2O$ may be emitted for high removal efficiencies with $SO_x$-rich gas.

Fly ash from coal has been used in Japan as a soil conditioner to supply plant micronutrients such as zinc and boron. Therefore, fly ash in small amounts in the product ammonium compounds may be accepted, but ash in large amounts lowers the fertilizer value.

Gas cooling using a water spray is simpler and gives higher $SO_2$ removal efficiencies than indirect cooling. The amount of water, however, must be controlled so that the water is entirely vaporized and the moisture content of the cooled gas does not exceed the critical humidity for the ammonium compounds. Use of excessive amounts of spray will cause moisture absorption by the compounds and produce wastewater which must be treated.

Ammonia leakage from the reactor may be reduced through improved control of the ammonia addition process. This involves measuring the concentration of outlet $NH_3$ as well as that of inlet $SO_x$ and $NO_x$. The formation of $N_2O$ may also be reduced but may not be entirely avoided. $N_2O$ is present in considerable concentrations in nature and small emissions do not have an adverse effect on the environment. However, further research on the reduction of $N_2O$ formation is needed. Table 6-1 shows that considerable portions of the byproducts are deposited in the duct. This deposition may be eliminated by increasing the gas velocity in the duct.

Although there are many simultaneous $SO_x$ and $NO_x$ removal processes, the Ebara process may be the only process commercially applicable to recover $SO_x$, $NO_x$ and $NH_3$ as useful byproducts. Further tests with a larger pilot plant using gases with various compositions are needed to prove the commercial applicability of the Ebara process.

## 6.4 ACTIVATED CARBON PROCESS

### 6.4.1 Introduction

The Electric Power Development Co. (EPDC), jointly with Sumitomo Heavy Industries, has conducted tests of the activated carbon process. The target is removal of 95% of the $SO_x$ and 40% of the $NO_x$ in the flue gas from a coal-fired boiler. A pilot plant with a capacity of treating 10,000 $Nm^3$/hr of flue gas from a coal-fired boiler at EPDC's Takehara Station was operated from November 1978 through December 1980.

With this process the $SO_2$ adsorbed by the carbon is regenerated by heating and is converted to elemental sulfur by the "RECLAUS PROCESS". This process combines the reduction of $SO_2$ to $H_2S$ and S with the reaction of $H_2S$ and $SO_2$ in a Claus furnace to obtain byproduct elemental sulfur. Ammonia is added to the flue gas for the following reasons: 1) to increase $SO_x$ removal efficiency, 2) to reduce carbon consumption by thermal regeneration, and 3) to remove a portion of the $NO_x$.

A demonstration plant with a capacity of treating 300,000 $Nm^3$/hr of flue gas from a coal-fired boiler at EPDC's Matsushima Station will be completed in 1984.

### 6.4.2 Description of Pilot Plant Test (21)

The composition of the flue gas and the test conditions during the pilot plant test are shown in Tables 6-4 and 6-5.

TABLE 6-4. COMPOSITION OF FLUE GAS

| | Range | Average |
|---|---|---|
| $SO_2$, ppm | 600-1500 | 1,000 |
| $NO_x$ ppm | 200-400 | 250 |
| $O_2$, % | 5.0-8.9 | 6.6 |
| $CO_2$, % | 12.0 | 12.0 |
| $H_2O$, % | 8.1-9.0 | 8.4 |
| Dust, mg/$Nm_3$ | 140-310 | 250 |

TABLE 6-5. TEST CONDITIONS

| | |
|---|---|
| $SO_2$, ppm | 700-1500 |
| NO, ppm | 200-350 |
| Gas temperature, °C | 120-150 |
| Space velocity, (SV) $hr^{-1}$ | 500-900 |

A flow sheet of the pilot plant is shown in Figure 6-10. The flue gas is added at about 0.7 mol ammonia to 1 mol $SO_x$ and passed horizontally through a moving bed in which granular activated carbon moves downward. $SO_x$ in the gas is adsorbed by the carbon to form sulfuric acid by the reactions:

$$SO_2 + H_2O + 1/2O_2 = H_2SO_4 \qquad (1)$$

$$SO_3 + H_2O = H_2SO_4 \qquad (2)$$

A portion of the ammonia reacts with sulfuric acid to form ammonium bisulfate while the rest of it reacts with NO by the catalytic action of the carbon:

$$NH_3 + H_2SO_4 = NH_4HSO_4 \qquad (3)$$

$$4NH_3 + 4NO + O_2 = 4N_2 + 6H_2O \qquad (4)$$

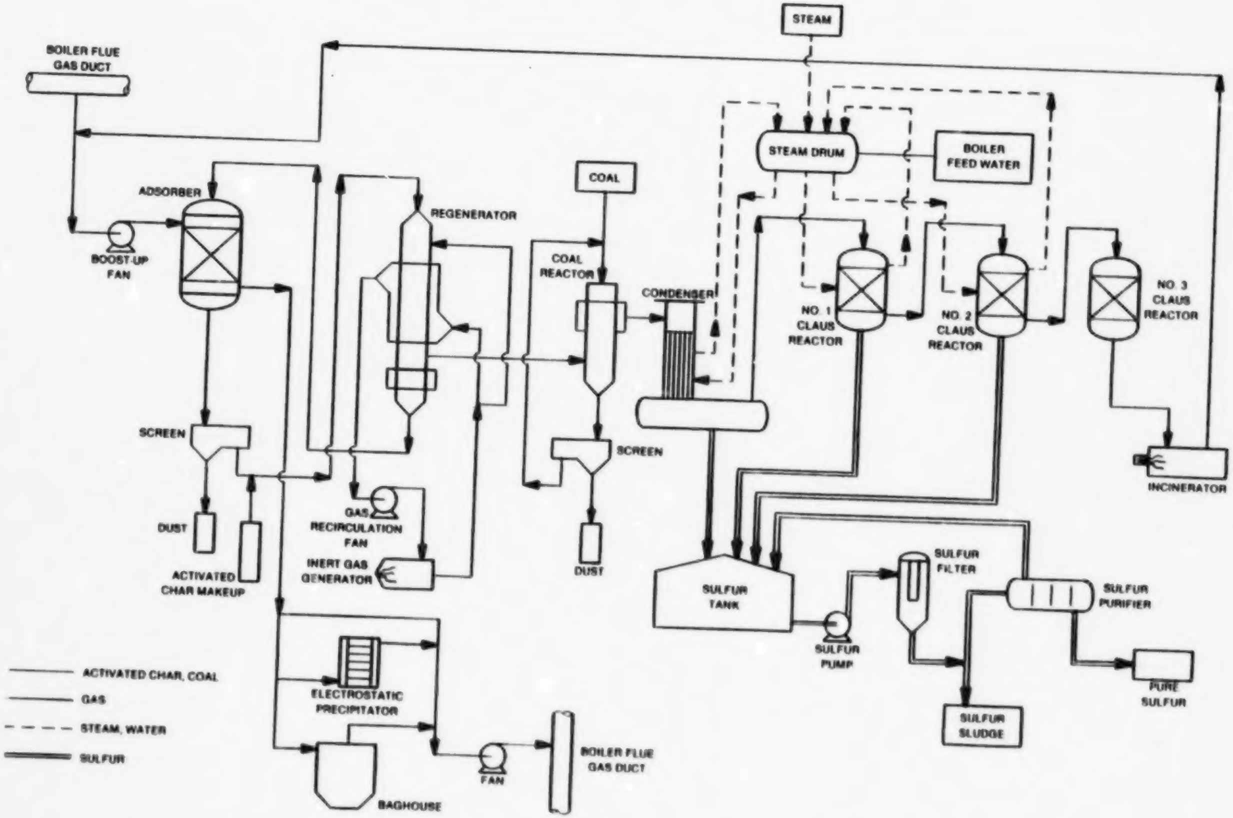

STEAM
BOILER FLUE
GAS DUCT
STEAM DRUM
BOILER
FEED WATER
COAL
ADSORBER
REGENERATOR
BOOST-UP
FAN
COAL
REACTOR
CONDENSER
NO. 1
CLAUS
REACTOR
NO. 2
CLAUS
REACTOR
NO. 3
CLAUS
REACTOR
SCREEN
SCREEN
INCINERATOR
GAS
RECIRCULATION
FAN
DUST
DUST
INERT GAS
GENERATOR
ACTIVATED
CHAR MAKEUP
SULFUR
FILTER
SULFUR
TANK
SULFUR
PURIFIER
SULFUR
PUMP
ACTIVATED CHAR, COAL
GAS
STEAM, WATER
SULFUR
ELECTROSTATIC
PRECIPITATOR
PURE
SULFUR
SULFUR
SLUDGE
BOILER FLUE
GAS DUCT
FAN
BAGHOUSE

Figure 6-10 Flowsheet of the Activated Carbon Process FGD Pilot Plant. EPDC's Takehara Station.

The relationship of reaction temperatures to removal efficiencies is shown in Figure 6-11.

The flue gas is then passed through an ESP or a baghouse and is led to the stack. The carbon, loaded with ammonium bisulfate and sulfuric acid, is discharged from the bed and introduced into another moving bed reactor (regenerator). There it is heated above 350°C by an inert gas produced by the incomplete combustion of fuel. $SO_2$ is regenerated by the following reactions:

$$2H_2SO_4 + C = 2SO_2 + 2H_2O + CO_2 \qquad (5)$$

$$2NH_4HSO_4 + H_2SO_4 = 3SO_2 + 6H_2O + N_2 \qquad (6)$$

The treated carbon is returned to the adsorber.

The $SO_2$-rich gas leaving the regenerator is mixed with a small amount of air and led to the bottom of a shaft furnace charged with coal. Due to the partial combustion of the coal, the gas temperature reaches about 900°C, resulting in the following reactions:

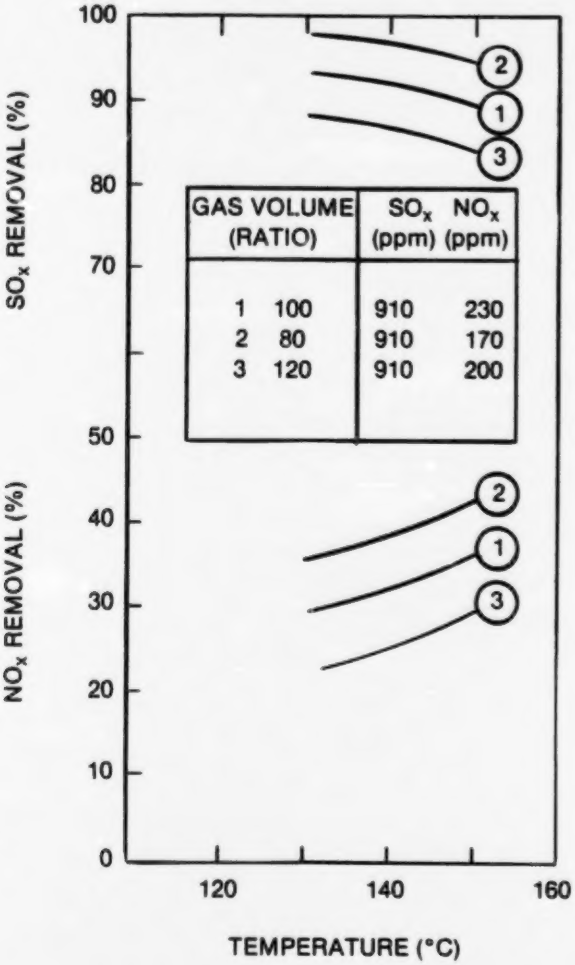
$C + H_2O = H_2 + CO$----(7) $\qquad$ $C + SO_2 = S + CO_2$----(9)

$H_2 + S = H_2S$----(8) $\qquad$ $CO + S = COS$----(10)

Approximately 90% of the $SO_2$ is reacted in the furnace where about 45% of it forms sulfur, 30% forms $H_2S$ and 10% forms COS. The gas is cooled to condense elemental sulfur, and is led into three Claus furnaces in series. In the furnaces, nearly all of the sulfur compounds are converted to elemental sulfur:

$$2H_2S + SO_2 = 3S \text{ and } 2H_2O \qquad (11)$$

$$COS + H_2O = H_2S + CO_2 \qquad (12)$$



| GAS VOLUME (RATIO) | SO$_x$ (ppm) | NO$_x$ (ppm) |
|---|---|---|
| 1 100 | 910 | 230 |
| 2 80 | 910 | 170 |
| 3 120 | 910 | 200 |

Figure 6-11 Effect of Temperature and Gas Volume on SO$_x$ and NO$_x$ Removal Efficiencies (NH$_3$/SO$_x$ = 0.7).

A portion of the $SO_2$-rich gas leaving the regenerator is fed into the Claus furnace for adjustment of the $H_2S/SO_2$ ratio. The gas leaving the Claus furnace contains small amounts of $H_2S$, $SO_2$ and COS. It is incinerated to convert $H_2S$ and COS to $SO_2$ and is returned to the adsorber.

Figure 6-12 shows the effect of the boiler load upon $SO_x$ and $NO_x$ removal efficiencies. At 100% load, the $SO_x$ and $NO_x$ removal efficiencies were 95-97% and 25-26%, respectively, while at 50% load the efficiencies were about 98 and 35%, respectively. Figure 6-13 shows the efficiencies during a continuous test period of 8,000 hours. The $SO_x$ removal efficiency ranged from 80 to 99% (90-95% removal most of the time) while the $NO_x$ removal efficiency ranged from 20-40%. The activated carbon consumption was 0.5 1% per day during the test period.

Ammonia was not detected in the flue gas leaving the adsorber as long as the $NH_3/SO_x$ mole ratio was kept below 0.8. The recovered sulfur contained a very small amount (below 0.5%) of impurities such as $NH_4HSO_4$ and $NH_4CL$. The impurities were removed by filtration to produce sulfur with a purity of over 99.9%.

### 6.4.3 Evaluation

The activated carbon process has several advantages. It does not require either water or gas reheating, removes over 90% of $SO_x$ simultaneously with about 30% of $NO_x$, and produces elemental sulfur. On the other hand, it consumes a considerable amount of carbon even with the addition of ammonia. The cost of the carbon may be one of the important factors which will affect the commercial applicability of the process.

The $NO_x$ removal efficiency of the process can be increased to 80-90% by using a much larger amount of ammonia and a high quality carbon at 230°C. This was demonstrated by the pilot plant testing of the Unitika Process(11). However, development of the Unitika process was abandoned because of the expense associated with the large amounts of carbon and ammonia consumed.

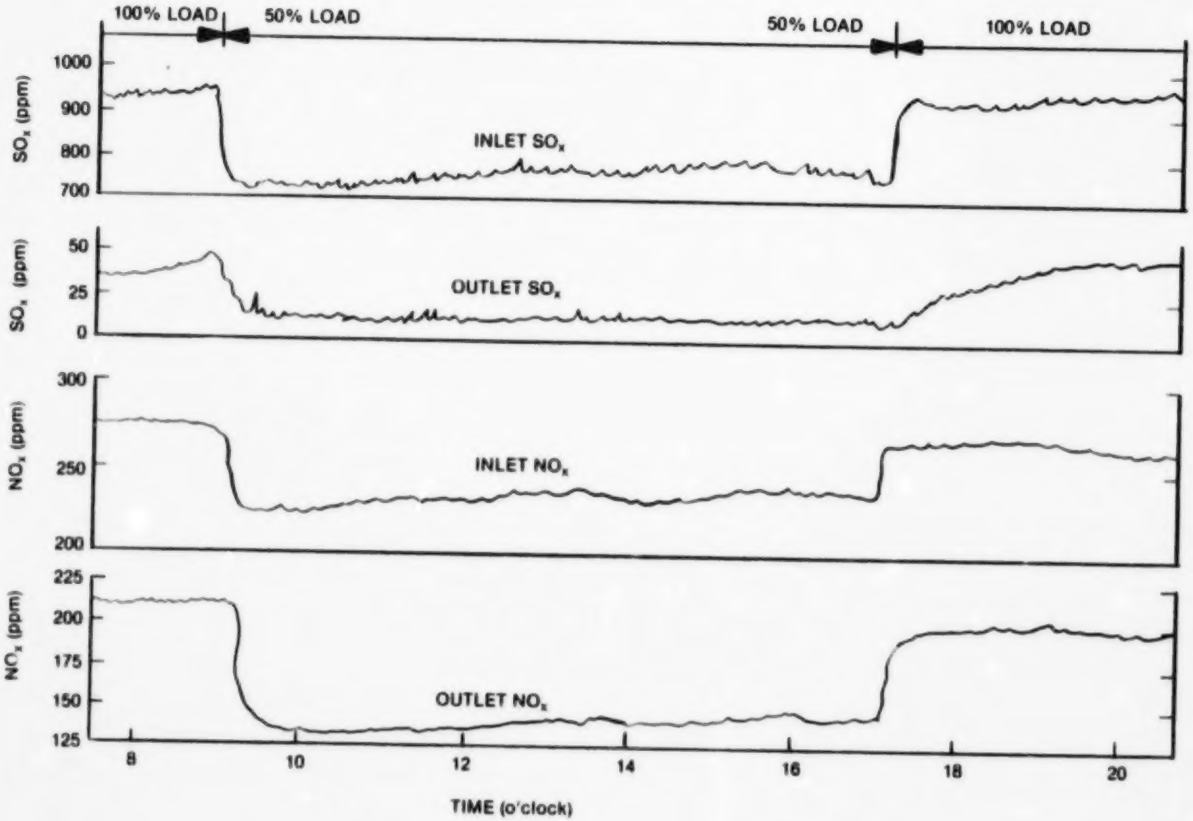


Figure 6-12 Effect of Boiler Load on $SO_x$ and $NO_x$ Removal Efficiencies (Activated Carbon Process).

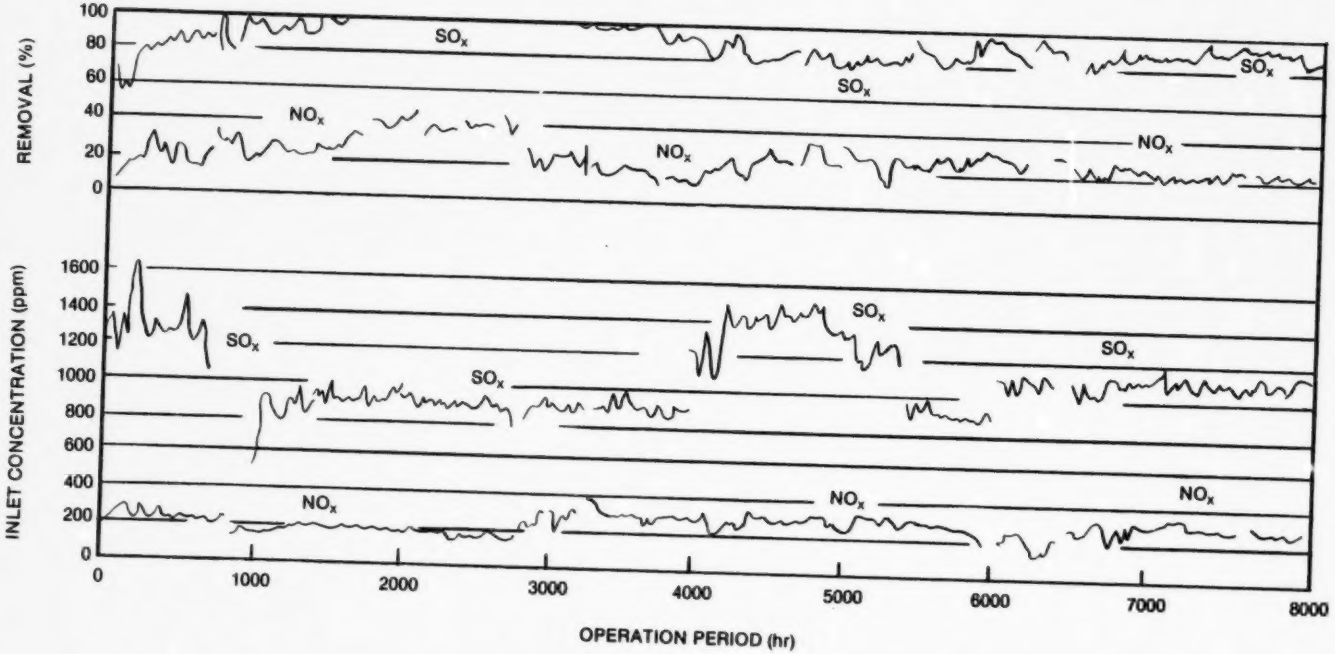


**Figure 6-13** SO$_x$ and NO$_x$ Concentrations and Removal Efficiencies During 8000 Hour Continuous Test of Activated Carbon Process.

The Unitika process may be more practical if inexpensive carbon and smaller amounts of ammonia are used for 30% $NO_x$ removal at lower temperatures.

During an early stage in the pilot plant testing of the activated carbon process, the RESOX process developed by Foster Wheeler, USA, was used for $SO_2$ reduction with coal. Since the conversion ratio remained about 60% with the RESOX process, the RECLAUS process has been developed based on a different concept to achieve about 90% conversion.

The activated carbon process works well with a flue gas that has a relatively low $SO_x$ content because carbon and ammonia consumption increase as the $SO_x$ content increases. The process might be able to compete with wet limestone FGD for coal or oil with less than 1% sulfur. The operation of the demonstration plant now under construction should provide more important information about the process.

## 6.5 ACTIVATED COKE SIMULTANEOUS $SO_x$ AND $NO_x$ REMOVAL

Mitsui Mining Co., the largest coal mining company in Japan, annually produces 2.4 million tons of coke, and has conducted tests of simultaneous $SO_x$ and $NO_x$ removal using activated coke. Mitsui Mining first studied the activated char process for $SO_x$ removal at the Luenen plant of Bergbau Forschung, West Germany, and modified the process to remove over 80% of the $NO_x$ and about 98% of the $SO_x$. Mitsui Mining has developed a process which uses a special coal treatment to produce activated coke suitable for simultaneous removal of $NO_x$ and $SO_x$. The coke does not contain metal components such as vanadium which act as $NO_x$ removal catalysts.

The simultaneous $NO_x$ and $SO_x$ removal pilot plant has a capacity of treating 1,000 $Nm^3$/hr of flue gas from a heavy oil-fired boiler at 150°C. Coal fly ash was added to the flue gas so that the effect of the ash could be studied. The pilot plant has two reactors in series; most of the $SO_2$ is adsorbed in the first reactor by the coke. A small portion of ammonia is fed to the first reactor, but the major portion is fed to the second reactor where most of the $NO_x$ is converted to $N_2$ and $H_2O$ by reacting with the ammonia. $SO_2$ is further adsorbed in the second reactor.

The adsorbed $SO_2$ is regenerated by a hot gas. The recovered concentrated $SO_2$ may be used for sulfuric acid or liquid $SO_2$ production. Mitsui Mining has conducted some studies on a method of producing elemental sulfur from the $SO_2$.

A demonstration plant with a capacity of treating 30,000 - 50,000 $Nm^3$/hr of flue gas from a coal-fired boiler may be constructed in 1983 to test this process further.

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